

CALIFORNIA INSTITUTE OF TECHNOLOGY

PROPERTY OF PHYSICS 34  
CALTECH

Library Number A 216

Circulation for research allowed.

☒ Reserve Copy. Not to be removed  
from 164 West Bridge.

Please Return To Rack II Shelf 3

**NORMAN BRIDGE LABORATORY OF PHYSICS**

**A POINT FOCUSING MONOCHROMATOR FOR**  
**THE STUDY OF LOW-ANGLE X-RAY DIFFRACTION**

**DETERMINATIONS OF THE SIZE DISTRIBUTIONS OF CARBON BLACKS  
AND A  
PRECISION DETERMINATION OF THE SIZE OF DOW LATEX PARTICLES**

by

**Leon Shenfil, Warren E. Danielson, and Jesse W. M. DuMond**

FILE COPY

PLEASE RETURN TO

Prof. Jesse W. M. DuMond  
Norman Bridge Laboratory of Physics  
California Institute of Technology  
Pasadena, California

**PHYSICS 34 LIBRARY**  
**DO NOT REMOVE**

**SPECIAL TECHNICAL REPORT NO. 15**

**A REPORT ON RESEARCH CONDUCTED UNDER  
CONTRACT WITH THE OFFICE OF NAVAL RESEARCH**

OCTOBER 1951

A POINT FOCUSING X-RAY MONOCHROMATOR FOR THE STUDY OF LOW  
ANGLE X-RAY DIFFRACTION. DETERMINATIONS OF THE SIZE  
DISTRIBUTION OF CARBON BLACKS AND A PRECISION DETERMINATION  
OF THE SIZE OF DOW LATEX PARTICLES.

PHYSICS 34 LIBRARY  
DO NOT REMOVE

CALIFORNIA INSTITUTE OF TECHNOLOGY  
Pasadena, California

Office of Naval Research  
Special Technical Report No. 15 on

FUNDAMENTAL RESEARCH IN SPECTROSCOPY  
OF SHORT WAVE-LENGTH X-RAYS AND GAMMA RAYS

Contract No. N6onr-244, T.O. IV  
NR 017-602

Dr. Jesse W.M. DuMond  
Contract Supervisor

A POINT FOCUSING X-RAY MONOCHROMATOR FOR THE STUDY OF LOW ANGLE  
X-RAY DIFFRACTION. DETERMINATIONS OF THE SIZE DISTRIBUTION OF  
CARBON BLACKS AND A PRECISION DETERMINATION OF THE SIZE OF DOW  
LATEX PARTICLES.

Special Technical Report No. 15  
October 18, 1951

Contract No. N6onr-244, Task Order IV  
California Institute of Technology  
Norman Bridge Laboratory of Physics

FUNDAMENTAL RESEARCH IN SPECTROSCOPY  
OF SHORT WAVE-LENGTH X-RAYS AND GAMMA RAYS

Contract Supervisor - Jesse W.M. DuMond

Foreword and Synopsis

This report describes a new instrument, the point-focusing x-ray monochromator, designed primarily for the study of low-angle x-ray diffraction phenomena whose principal application, we believe, will be for the determination of the sizes and other features of small particles and large organic molecules of the order of say a hundred up to a few thousand angstroms diameter in the range of sizes below and up to those studied in present day electron microscopy. This diffraction method, in contrast to electron microscopy, possesses the disadvantage that it is less direct and does not reveal information about individual particles. On the other hand, it does permit the study of wet samples which cannot be studied with the electron microscope. In many cases, the nature and geometry of the particles or molecules of interesting organic substances are so radically changed by the dessication process necessary to prepare them for examination with the electron microscope, that this instrument can reveal little about such particles or molecules as they really are in their natural state. This seems apt to be particularly true of many long chain molecules of interest to the biochemist. The value of a method which permits obtaining information regarding wet samples is thus obvious.

Up to the present, the art of low-angle x-ray diffraction, with a few exceptions, has been largely restricted to studies with



radiation only roughly monochromatized by means of filters and collimated with fine slits or pinholes. Where slits were used of such dimensions that the primary (unscattered) beam formed a line rather than a point image, (to give somewhat more intensity) a severe difficulty of interpretation entered if the line was of the same order of length as the diffraction pattern to be studied (a requisite in order to secure a substantial gain in intensity) since the 1 to 1 correspondence between points on the diffraction pattern and diffraction angles was thereby destroyed. Collimation with pinholes or slits fine enough to permit really high angular resolution (say such as to limit the primary beam within an angular range of a minute or so of arc) had to be accomplished at a severe sacrifice of intensity. Fogging in the vicinity of the primary beam due to scattering from the slit system was also difficult to suppress.

In an effort to overcome some of these difficulties the supervisor of this contract conceived the idea in 1949 of using two successive focusing curved crystal reflections, the two focusings to occur in two planes mutually at right angles so as to achieve a monochromatic point focused x-ray beam without astigmatism by means of a special geometrical disposition of the crystals and of their reflecting atomic planes. This fundamental geometry was disclosed in a brief Letter-to-the-Editor of the Review of Scientific Instruments (Fig. 1, p. 188) in the February 1950 issue of that journal. Work on the development of a practical realization of these ideas was postponed for lack of funds however until September 1950 when authorization for the inclusion of this development under Office of Naval Research Contract, N6onr-244, T.O. IV permitted construction work to be started.

The physical design of the point focusing monochromator, its construction, the difficult and exacting profiling of the stainless steel clamping blocks which impart the correct curvature to the two monocrystalline quartz laminae, and the elaborate adjustment of the parts by a carefully planned procedure to give an anastigmatic point-focused primary beam are entirely the result of the joint effort of two able graduate students, Leon Shenfil and Warren E. Danielson of this Institute. The difficult and skillful analysis (by tracing a general ray through the entire system from the x-ray tube target through the two successive focusing curved crystal reflections to the point focus on the photographic film taking into account the inhomogeneity in wave-length in the natural breadth of the  $\text{CuK}\alpha_1$  x-ray line and the slight departures of the diffraction angle for reflection at different points on the surfaces of the two focusing curved crystals) is the original work of L. Shenfil and constitutes part of his doctorate thesis. By this analysis, he has succeeded in explaining correctly and quantitatively the shape and small but finite size of the point focus.

Sharp point foci of the primary beam, remarkably free from surrounding fog when no scattering sample is interposed, have been obtained. Although, the focus is anastigmatic as regards focusing



by the two crystals the spot is slightly elongated in a direction approximately at  $45^\circ$  to the astigmatic axes defined by the two crystal reflections for a rather involved reason which Shenfil's analysis explains quantitatively. The shape of this tiny elongated focal image spot is in fact an image of the (projected) utilized area of the focal spot on the x-ray tube target (the area projected on a plane normal to the emerging beam). Along the length of this elongated spot, Shenfil has shown that the wave-length of the radiation varies over the range comprised in the natural breadth of the characteristic x-ray line employed (in this case  $\text{Cu K}\alpha_1$ ). By limiting the projected useful area of the x-ray tube focal spot by means of adjustable stops this image focal spot can be further reduced in size but at some expense as to intensity. Without any such limitation in the present instrument the width of the elongated primary image on the photographic film (minor axis of the spot) subtends only about  $1\frac{1}{3}$  minutes of arc at the position of the scattering sample when this latter is at the greatest possible distance from the photographic film (i.e., adjacent to the second crystal holder) and in the absence of a scattering sample, the fogging immediately adjacent to this narrow spot is almost entirely nil even after very prolonged exposures. The long dimension of the unrestricted spot is only about six times its width. These figures give an idea of the very satisfactorily high angular resolving power attainable with a sharply monochromatic beam.

The study of particle sizes of certain carbon black samples of heterogeneous size distribution also constitutes part of L. Shenfil's thesis, which is here reproduced in toto as the first and longest part of the present Special Technical Report.

Mr. Warren Danielson, whose thesis, as yet unfinished, will consist in part of studies of various substances with the instrument which he has collaborated in constructing, has written a brief preliminary account for this report (following the part by Mr. Shenfil) describing measurements recently made with the point-focusing x-ray monochromator on the diameters of the now famous Dow Chemical Co., Lot 3584, Batch 580-G Latex particles. These particles which for some mysterious unexplained reason proved upon electron microscope examination to consist of spherical particles extremely uniform in size have proven of great value to microscopists in many quarters of the world as a standard of comparison, and accordingly the establishment of their mean diameter in absolute units with good precision and freedom from systematic error is a matter of considerable and widespread interest. The instrument appears capable of yielding a precision in such measurements of the order of one tenth percent. Reproductions of a few of the concentric ring diffraction patterns from these spherical particles are shown (p. 90). Much more work on this problem (as briefly indicated in Mr. Danielson's report) still remains to be done and the present preliminary results are given merely as an indication of the possibilities of the new instrumental techniques.

In particular, it is hoped that considerable gain in intensity over that at present available can be effected by several modifications

suggested in Mr. Shenfil's thesis (p. 44) and it is hoped that Mr. Danielson may be able to incorporate the realization of some of these improvements as part of his thesis work for the doctorate.

The undersigned wishes to take this opportunity to express his gratitude to the Office of Naval Research for their generous permission to include financing of this development under Contract N6onr-244, T.O. IV. He also wishes to thank Prof. L. Pauling for his encouragement and supporting recommendation of the proposal to the O.N.R. when the primary idea was in a nascent stage. He is very deeply indebted to the two graduate students whose patience, skill and diligence have so successfully brought the initial ideas to concrete realization in spite of many difficulties and uncertainties.

Respectfully submitted,

*Jesse W.M. DuMond*

Jesse W.M. DuMond

POINT FOCUSING X-RAY MONOCHROMATOR

FOR

LOW ANGLE SCATTERING STUDIES

Thesis by

Leon Shenfil

In Partial Fulfillment of the Requirements

For the Degree of

Doctor of Philosophy

California Institute of Technology

Pasadena, California

1952



### ACKNOWLEDGEMENTS

I wish to take this opportunity to thank Professor Jesse W.M. DuMond for suggesting this thesis project and for taking a keen interest in the course of its development. His helpful suggestions have been instrumental in the success of this work. In the three years that I have worked for him as a graduate student, I have profited considerably from his vast experience and have learned a good deal of the practical side of physics, the sort of thing one does not find in textbooks. For this, I wish to express my deepest gratitude.

To Warren E. Danielson, who shares equally in the design and construction of the point focusing monochromator, I wish to extend my hearty thanks and the hope that he will find many interesting applications of the instrument.

This project was supported jointly by the Office of Naval Research and the Atomic Energy Commission.

## ABSTRACT

A point focusing x-ray monochromator was designed and constructed for low angle scattering studies. The anastigmatic point focus is achieved by means of two cylindrically bent quartz crystals whose focal circles are mutually perpendicular. The beam, emanating from the copper target of an x-ray tube, is reflected in succession, first from the crystal defining the horizontal focal circle and, second from the crystal defining the vertical focal circle following which it comes to a monochromatic point focus of wavelength  $1.537 \text{ \AA}$  ( $\text{Cu K}\alpha_1$ ). The sample to be studied is placed between the second crystal and the point focus, and the scattered beam is detected by means of a photographic plate placed at the point focus, at right angles to the undeviated beam, the latter being suppressed by means of an absorber.

Mathematical analysis, in which a ray was traced through the two crystal system, revealed correctly the shape and size of the point focus, and the possibility of reducing the latter in size by stopping down the beam emerging from the target.

The instrument and its lining up procedure is described in detail, particularly the latter since the orientation of the two crystals relative to each other and the x-ray tube involve twelve degrees of freedom.

Suggestions for improving the intensity of the instrument are made. These consist of (1) substitution of topaz crystals for quartz, (2) use of helium atmosphere instead of air to surround the x-ray beam (3) reorientation of the x-ray tube to permit smaller angles of emergence of the beam.

As a trial run for the instrument, the scattering patterns of two kinds of carbon black (good scatterers) were obtained from which the average particle sizes and size distributions were calculated.



# TABLE OF CONTENTS

<u>PART</u>	<u>TITLE</u>	<u>PAGE</u>
I	INTRODUCTION	1
II	GEOMETRY OF THE POINT FOCUSING MONOCHROMATOR	6
	Geometry of a Single Bent Crystal	6
	Tracing a General Ray through the System	11
	Aberrations	34
	Table of Nomenclature (for Part II only)	39
III	THE INTENSITY PROBLEM AND SUGGESTED IMPROVEMENTS	44
IV	DESCRIPTION OF THE POINT FOCUSING MONOCHROMATOR	53
	The Crystals and Crystal Blocks	63
V	EXPERIMENTAL RESULTS	68
	Method of Detection	69
	Low Angle Scattering Due to Small Particles	72
	Particle Size Determinations of Carbon Blacks	77
	REFERENCES	81

PART I

INTRODUCTION

In the past few years a good deal of interest has arisen in low angle x-ray diffraction studies, particularly among chemists and biologists. It is well known<sup>(1)</sup> that x-rays scattered at low angles reveal the size, and possibly the shape of small particles that make up the scatterer. In the case of fibrous substances, such studies<sup>(2)</sup> may reveal the size of large periodicities along the fibers. In a way, one can think of low angle studies as being an extension of microscopy into smaller sizes although the "pictures" that one obtains are by no means as easy to interpret. In the case of particles, the intensity, in general, is a function of  $\epsilon r/\lambda$  where  $\epsilon$  is the scattering angle,  $r$  the particle radius and  $\lambda$  the x-ray wavelength being scattered.<sup>(1,3)</sup> Theoretical curves of intensity versus scattering angle have been computed for particles of various sizes, shapes and size distributions<sup>(4)</sup>, so that all one has to do is to match the intensity distribution of the scattered x-rays with one of the theoretical curves in order to determine the physical properties of the scatterer. This seemingly simple procedure, however, is complicated by several factors. The intensity distribution that one obtains in practice is the fold (or convolution) of the spectral distribution of the x-rays, the beam aperture, and the scattering distribution, thus posing the problem of separating or unfolding the first two factors in order to obtain the actual scattering distribution. The problem of unfolding is a very difficult one particularly

if one doesn't know exactly what functions formed the fold in the first place.

Thus, to obtain a scattering pattern which is truly representative of the scattering medium, one has to try to minimize the instrumental effects, by constructing an instrument which will produce a monochromatic beam of high spectral resolution. However, as is usually the case, the price of high resolution is loss of intensity which in many cases is so serious as to defeat the whole purpose of the experiment. Various types of instruments have been built in the past in which a compromise, of some form or another, has been made between high resolution and low intensity. These instruments can be classed roughly into two types: (1) the slit type instrument consisting of an x-ray beam, possibly filtered, which is collimated by a slit system or (2) the crystal type, in which one or two crystals (either flat or bent) are used to monochromatize the beam.

The slit type instrument is best adapted for long wavelengths, thus requiring vacuum methods. This is a handicap which is shared by an electron microscope, especially when scattering from liquid samples is desired. Successful use of such an instrument has been reported by Yudowitch<sup>(5)</sup>, and Bolduan and Bear<sup>(2,6)</sup>.

An instrument for low angle scattering studies in which a curved crystal is used for monochromatization has been reported by Guinier<sup>(1)</sup> and later at the suggestion of DuMond,



has been modified by the addition of a second curved crystal placed in the "parallel"\* position with respect to the first.\*\* See Fig. 1. In the latter scheme, the sample is placed in the convergent beam, midway between the second crystal and the line focus, while the photographic plate is placed in the focal plane. The diffraction pattern appears on the photographic plate as a diffuse distribution symmetrically disposed on either side of the direct beam, the latter being suppressed by means of an absorber. It has been found that line focused beams are not well adapted in the study of the long chains of protein molecules in which the periodicities of interest are along the chain.

The possibility of bringing a monochromatic beam of x-rays to an anastigmatic point focus was first suggested by DuMond in 1949<sup>(7)</sup>, however, due to lack of funds the design and construction of a point focusing instrument were not undertaken until the summer of 1950.

---

\*The "parallel" position is the position of zero dispersion. See Compton and Allison (8), p. 718, where the term is used in relation to two flat crystals.

\*\*The beam is focused monochromatically to a line by reflection from the first crystal and is refocused again to a line by reflection from the second crystal, thus cutting down the incoherent scattering in the neighborhood of the line focus. This is not to be confused with the instrument whose development forms the subject matter of the present thesis in which the angles of deviation of the beam in the two successive crystal reflections lie in planes at right angles to each other in such a way that the beam is brought to an anastigmatic point focus.

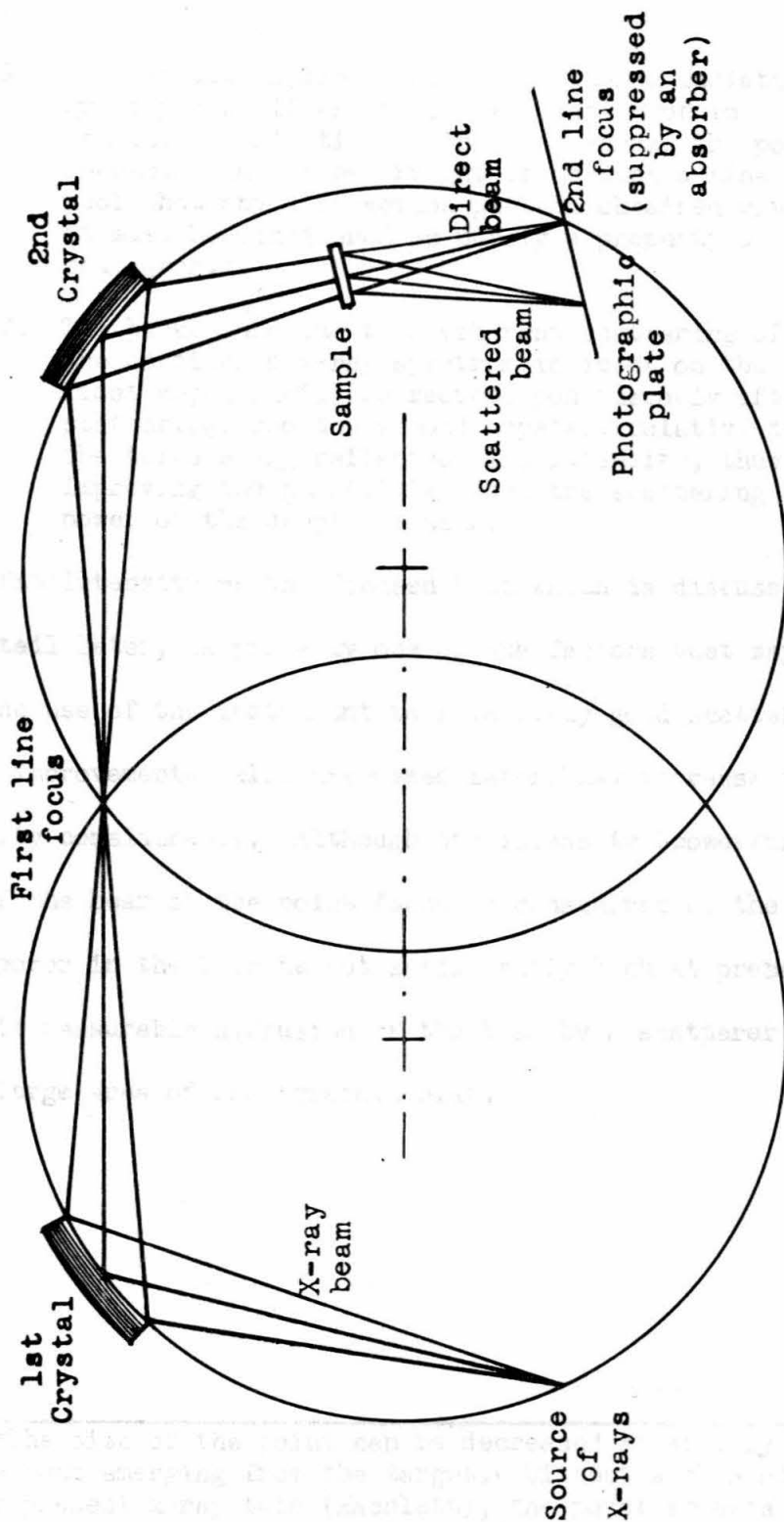


Fig. 1 - Two crystal line focusing monochromator.

The advantages of a point focusing monochromator are two-fold:

1. The circular symmetry of the scattered radiation greatly simplifies the interpretation of intensity distributions. In other words, the point focused beam may be thought of as such a fine tool that any diffraction pattern obtained with it must be considered as purely a property of the scatterer.\*
2. The background due to incoherent scattering of the continuous x-ray spectrum incident on the first crystal will be reduced considerably after scattering from the second crystal, relative to the twice Bragg reflected line intensity, thus improving the sensitivity when the scattering power of the sample is weak.

The intensity of the focused beam which is discussed in some detail later, is probably one of the factors that may limit the use of the instrument to relatively good scatterers. Several improvements, also discussed later, may increase the luminosity considerably. Although the intensity (power/unit area) of the beam at the point focus is considerable, the actual power in the beam is not sufficiently high at present to permit measurable diffusion of the beam by a scatterer over a large area of photographic plate.

---

\*The size of the point can be decreased at will by stopping down the beam emerging from the target. Without such stops and with our present x-ray tube (Machlett), the point appears slightly elongated with dimensions 0.196 by 1.40 mm.



## PART II

### GEOMETRY OF THE POINT FOCUSING MONOCHROMATOR

Since the point focusing monochromator consists of two bent crystals, each of which behaves like a line focusing monochromator, it will be well to review briefly the geometry of a single bent crystal. The numerous symbols used in the discussion that follows are listed alphabetically at the end of this part (p. 39).

#### GEOMETRY OF A SINGLE BENT CRYSTAL

It is well known<sup>(9)</sup> that x-rays can be made to focus monochromatically by a crystal lamina which has been hollowed out, cylindrically, to a given radius and then bent elastically to half that radius. If we let the concave face of the crystal after bending define a cylinder, then the intersection of this cylinder with a plane, bisecting the crystal and perpendicular to the generators, is a circle called the focal circle. The plane is called the focal plane. The only condition required for point focusing is that the line, about which the crystal planes are concentric, be one of the generators of the focal cylinder.

Fig. 2 shows a single crystal with its focal circle. C is the center of the circle, S a point-source of x-rays, F the vertical line focus, P some arbitrary point on the crystal, and  $\mathcal{B}$  the vertical line about which the crystal planes are concentric. Thus, the figure shows the projection of the rays  $\overline{SP}$  and  $\overline{PF}$  on the focal plane with  $\overline{\mathcal{B}P}$  normal to the crystal



planes at P. ( $\overline{SP}$  and  $\overline{PF}$  are in a plane not necessarily parallel to the focal plane.) If the arcs  $\widehat{SB}$  and  $\widehat{BF}$  are equal, then the angle of incidence  $\widehat{SPB}$  is equal to the angle of reflection  $\widehat{BPF}$ . This relation holds true for all points on the crystal surface and, furthermore, if the vertical divergence (angle between rays  $\overline{SP}$  or  $\overline{PF}$  and the focal plane) is not excessive, the angle of incidence remains fairly constant for all points on the crystal surface. The above two conditions are precisely the ones required for reflection of monochromatic x-rays. The wavelength reflected is given by the Bragg law  $\lambda = 2d \sin \theta$  where  $\theta$  is the grazing angle which is also the complement of the angle of incidence. Thus, it has been shown that a beam emanating from the point S will focus monochromatically at the vertical line through F. Experimentally, the position of point S, relative to the crystal, is such that the  $K\alpha_1$  line of Cu is reflected in the monochromator here described.

In Fig. 2, the point on the focal circle diametrically opposite B, is denoted by B', and the center of the crystal lamina by A. It should be clear from this figure that a much larger crystal lamina (such as that indicated, partly with lighter lines, between the points ZZ in the figure), with center at B' could be used to form the monochromatic image at F of the source S. In the present instrument only a portion ZZ' entirely on one side of B' is actually used. The reason why the center of the crystal is not at B', but to the left of it, will be evident as soon as the two-crystal arrangement is

discussed.

The angle  $2\alpha$  is a measure of the angular displacement of point A from  $S'$ . Since the angle  $\widehat{SAC}$  is equal to  $\alpha$ , then  $\alpha$  is also the dihedral angle between the crystal plane and its face (or between their normals) at the point A and at all points lying on the generator through A.

The reflected beam that arrives at F appears to come from the virtual line source F' which lies on a circle of radius  $SS'$  with  $S'$  as center, see Fig. 3. Now let us take a second identical crystal with its focal plane perpendicular to the focal plane of the first crystal and place it in such a way that the line of intersection of the two focal planes is  $FF'$ . Let the center of the second crystal be denoted by B, then both B and A are on the line  $FF'$ . If we let  $BF' = AF$  and if the point F' lies on the focal circle of the second crystal, then the rays striking this crystal will be reflected at the Bragg angle and will come to a point focus, P.F.

It should now be evident, (1) why only a part,  $ZZ'$ , of the total conceivable crystal  $ZZ$  shown in Fig. 2 is retained, (2) why the center A of the crystal  $ZZ'$  is displaced angularly by an amount  $2\alpha$  from the point  $S'$  and (3) why the atomic reflecting planes of the crystal are not exactly parallel to the face of the bent crystal slab at its center but make an angle  $\alpha$  therewith. All three of these requirements originate from the fact that the first crystal must be kept entirely on one side of the point  $S'$  and the second crystal entirely on

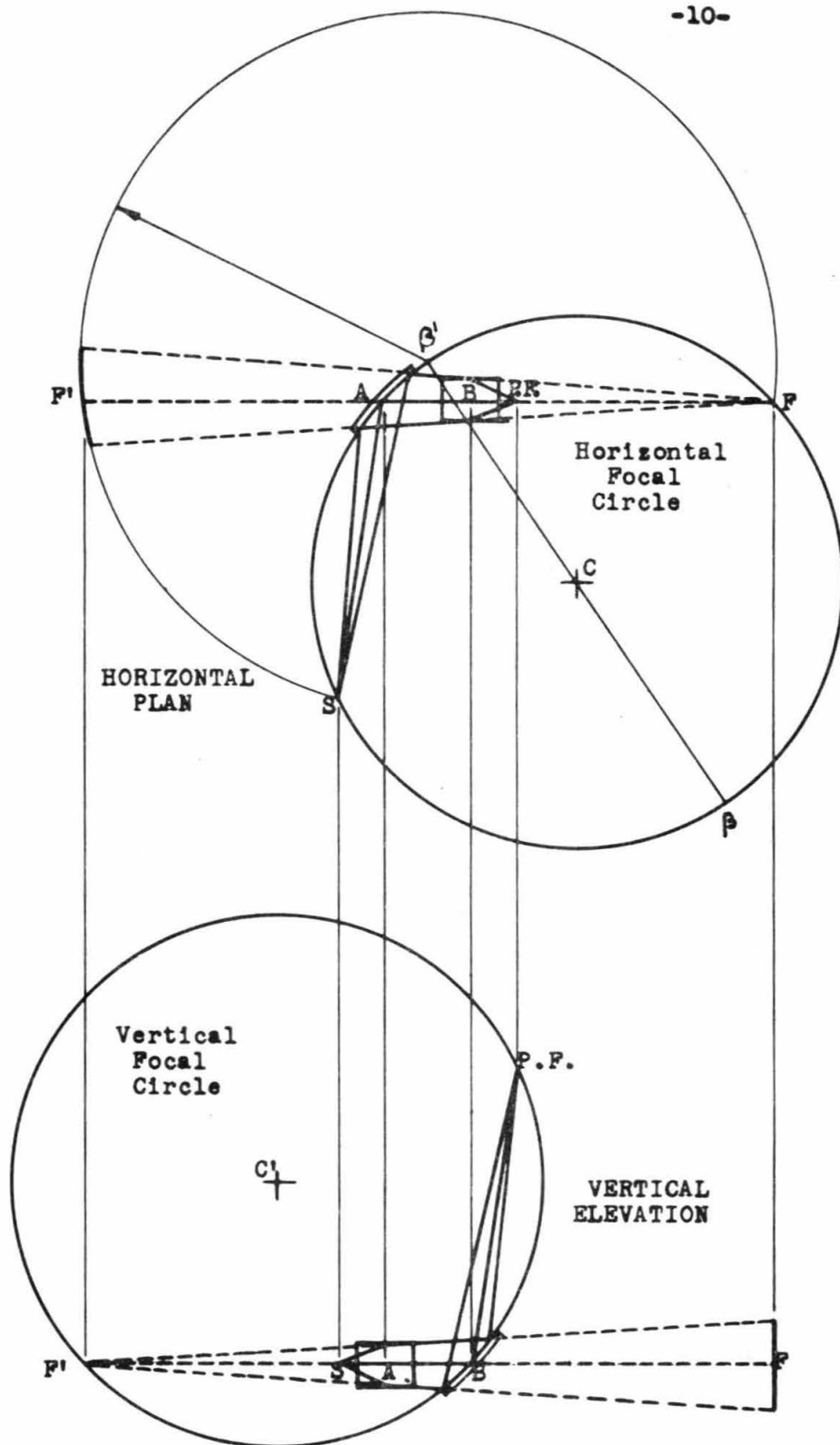


Fig. 3 - Two bent crystals arranged to form a point focusing monochromator.

the other side so as to avoid interference between the two crystals or their crystal holders when the two focal circles are correctly mounted to avoid astigmatism. Clearly, the larger the angle  $\alpha$  is made, the farther apart the two crystal clamping blocks may be placed. The distance between the centers A and B of the two crystals is given by  $4r \sin \alpha \cos \theta$  where  $r$  is the radius of either focal circle.

If the two focal circles were in the same plane and superimposed on one another (without turning over), with point B coinciding with A, and with point focus (P.F) coinciding with the point source (S) then the virtual source (F'), if we neglect the slight curvature, would coincide with the line focus (F). It is thus seen that the point focus monochromator is made up of two identical line focus monochromators placed in a definite way relative to one another.

In order to examine the aberrations that occur in such a point focusing arrangement, it is necessary to take a general ray and trace it through the system.

#### TRACING A GENERAL RAY THROUGH THE SYSTEM

For convenience the unit of length is taken equal to the radius of one of the focal circles. Let the coordinates describing the horizontal focal circle be  $x, y, z$ , or when convenient, cylindrical coordinates  $\rho, \phi, z$ , where  $z$  is perpendicular to the focal plane and is also measured in units of  $r$  and  $\rho$  is the radial distance from the axis passing through C normal to the focal circle.



In Fig. 4,  $S_0$  represents the center of the target which is on the focal circle,  $S$  the intersection of a ray from an arbitrary point on the target with the focal cylinder\*,  $A$  the center of the crystal,  $P$  some arbitrary point on the crystal with coordinates  $(1, \phi, z)$ ,  $F_0$  the focal point, and  $\beta$  the vertical axis about which the crystal planes are concentric. The x-axis is in the focal plane, perpendicular to  $\overline{AF}$ . In the calculations that follow, we will derive the relation between the grazing angle  $\theta$ , defined on p. 14 and the Bragg angle  $\theta_0$ , defined below.

$\overline{S_0P}$  represents a ray of wavelength  $\lambda_0$  such that the angle  $\theta_0$  satisfies the Bragg law  $\lambda_0 = 2d \sin \theta_0$ .  $\lambda_0$  is the wavelength at the middle of the  $K\alpha_1$  line.

The angle between the projection of  $\overline{S_0P}$  on the xy plane and the x-axis is  $-(\frac{\pi}{2} - \frac{3\theta_0 - \alpha + \phi}{2})$ .\*\* The length of  $\overline{S_0P}$  is

$$\begin{aligned} |\overline{S_0P}| &= \left[ 4 \sin^2 \left( \frac{3\theta_0 - \alpha - \phi}{2} \right) + z^2 \right]^{1/2} \\ &= 2 \sin \frac{3\theta_0 - \alpha - \phi}{2} \left[ 1 + z^2 / (4 \sin^2 \frac{3\theta_0 - \alpha - \phi}{2}) \right]^{1/2} \end{aligned}$$

and the length of its projection on the xy plane is

$$2 \sin \frac{3\theta_0 - \alpha - \phi}{2}$$

---

\*The plane of the target is not tangent to the focal cylinder. It is oriented in such a way that the central ray  $\overline{S_0A}$  makes approximately  $6^\circ$  with it. This direction of  $\overline{S_0A}$  is the direction of maximum intensity.

\*\*The angle  $\alpha$  has the same significance explained for Fig. 2 and the angle  $\phi$  in Fig. 4, is the angle  $\widehat{PCX}$ .

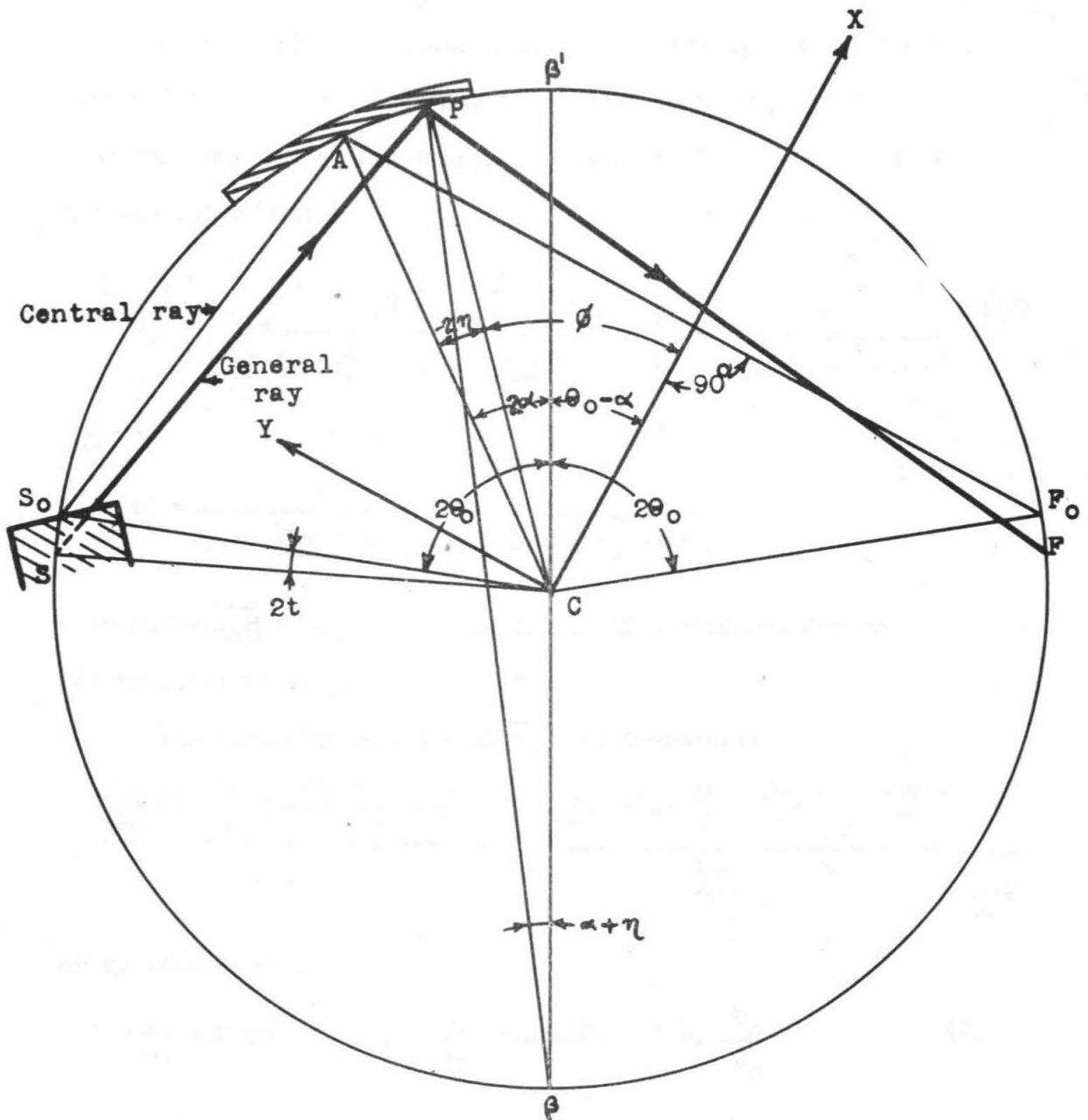


Fig. 4 - Horizontal focal circle shown with its coordinate axes.

It will be convenient to measure angles from  $\overline{AC}$  rather than the x-axis. We therefore define the central angle  $\widehat{ACP}$  by  $2\eta$  and from Fig. 4 it is seen that,

$$2\eta = \phi - \theta_0 - \alpha \quad (1)$$

Because of the limited size of the crystal,  $\eta$  will never exceed  $1/80$  radians. This makes it possible to neglect terms of order  $\eta^3$ , since they represent angles that are smaller than two seconds of arc.\*

Let,

$$a_0 = \left[ 1 + \frac{z^2}{4\sin^2 \frac{3\theta_0 - \alpha - \phi}{2}} \right]^{1/2} \approx \left[ 1 + \frac{z^2}{4\sin^2 (\theta_0 - \alpha - \eta)} \right]^{1/2}$$

and

$$b_0 = \frac{z}{2\sin \frac{3\theta_0 - \alpha - \phi}{2}} = \frac{z}{2\sin (\theta_0 - \alpha - \eta)}$$

This gives  $|\overline{S_0P}| = za_0/b_0$  and the length of its projection on the xy plane is  $z/b_0$ .

The direction cosines of  $\overline{S_0P}$  are therefore:

$$\frac{\frac{z}{b_0} \cos(\frac{\pi}{2} - \frac{3\theta_0 - \alpha + \phi}{2})}{\frac{za_0}{b_0}}, \quad \frac{\frac{z}{b_0} \sin(\frac{\pi}{2} - \frac{3\theta_0 - \alpha + \phi}{2})}{\frac{za_0}{b_0}}, \quad \frac{z}{\frac{za_0}{b_0}}$$

or by substituting (1)

$$\frac{1}{a_0} \sin(2\theta_0 + \eta), \quad -\frac{1}{a_0} \cos(2\theta_0 + \eta), \quad \frac{b_0}{a_0} \quad (2)$$

---

\*The full-width at half maximum of the Darwin diffraction pattern for  $K\alpha_1$  radiation of copper reflected from quartz in the 1st order of the (310) planes is about 2 seconds of arc and this, therefore, has been taken as the limit of negligibility.

The above ray is emitted from the point  $S_0$  whose coordinates are  $(1, 3\theta_0 - \alpha, 0)$ . Suppose now, a ray is emitted from the neighborhood of  $S_0$ , say the point  $S$ , whose coordinates relative to  $S_0$  are  $(t_y, t_z)$ , see Fig. 5, or relative to the focal circle  $(1, 3\theta_0 - \alpha + 2t, t_z)$  where  $t = t_y/2\sin(\theta_0 - \alpha)$ , then the direction cosines of  $\overline{SP}$  follow from (2)

$$\frac{1}{a} \sin(2\theta_0 + \eta + t), \frac{1}{a} \cos(2\theta_0 + \eta + t), \frac{b}{a} \quad (3)$$

where

$$a = \left[ 1 + \frac{(z - t_z)^2}{4 \sin^2(\theta_0 - \alpha - \eta + t)} \right]^{1/2} \quad (4)$$

and

$$b = \frac{z - t_z}{2 \sin(\theta_0 - \alpha - \eta + t)} \quad (5)$$

The projected target size is 1 mm. x 1 mm. at right angles to the central ray so that  $|t| = |t_y/2\sin(\theta_0 - \alpha)| \leq 1/1500$  radians and  $|t_z| \leq 1/1200$  radians.

$\overline{PB}$  represents the normal to the crystal planes at point P and its direction cosines are:

$$\cos \frac{\phi + \theta_0 - \alpha}{2}, \sin \frac{\phi + \theta_0 - \alpha}{2}, 0$$

or by substitution of (1)

$$\cos(\theta_0 + \eta), \sin(\theta_0 + \eta), 0 \quad (6)$$

Now let  $\theta$  be the grazing angle of the ray at point P, i.e. the angle that the ray makes with the crystal planes, so

that  $\frac{\pi}{2} - \theta$  is the angle between  $\overline{SP}$  and  $\overline{PB}$ , then

$$\cos\left(\frac{\pi}{2} - \theta\right) = \sin \theta = \frac{\overline{SP} \cdot \overline{PB}}{|\overline{SP}| |\overline{PB}|}$$

With the help of (3) and (6), this gives

$$\sin \theta = (1/a) \left[ \sin(2\theta_0 + \eta + t) \cos(\theta_0 + \eta) - \cos(2\theta_0 + \eta + t) \sin(\theta_0 + \eta) \right]$$

or

$$\sin \theta = (1/a) \sin(\theta_0 + t) \quad (7)$$

From (4) and (5),  $a = (1 + b^2)^{1/2}$ , where  $b^2$  is small, so that terms of order  $b^4$  in the expansion of  $1/a$  can be neglected.

Therefore,

$$1/a \approx 1 - b^2/2 \quad (8)$$

and

$$\sin \theta \approx \sin(\theta_0 + t) - (b^2/2) \sin(\theta_0 + t)$$

The second term on the right represents a small correction to the angle  $\theta$ , which can be expressed as follows:

$$\theta = \theta_0 + t - \gamma \quad (9)$$

$$\sin \theta = \sin(\theta_0 + t - \gamma) \approx \sin(\theta_0 + t) - \gamma \cos(\theta_0 + t)$$

$\gamma$  is identified by comparing with the preceding equation, and is given by

$$\gamma = \frac{b^2}{2} \tan(\theta_0 + t) \quad (10)$$

or upon elimination of  $b$  with equation (5), by

$$\gamma = \frac{(z - t_z)^2 \tan(\theta_0 + t)}{8 \sin^2(\theta_0 - \alpha - \eta + t)} \approx \frac{z^2 \tan \theta_0}{8 \sin^2(\theta_0 - \alpha)} \quad (10a)$$

Also, since  $z$  is at most  $1/60$  radians and the arguments of the tangent and sine are approximately  $\pi/4$  so that  $\gamma \approx 15$  sec. of

arc, it was therefore justifiable to omit terms of order  $b^4$  in eq. (8).

We have thus obtained an expression for the grazing angle  $\theta$  of a ray of wavelength  $\lambda_0$ , from the point S, in terms of the Bragg angle  $\theta_0$  and the small angular deviations  $t$  and  $\gamma$ .

It is now necessary to consider wavelengths that are slightly different from  $\lambda_0$ , since the whole  $K\alpha_1$  line contributes to the beam rather than just a small portion of it represented by  $\lambda_0$ . Let the new wavelength be  $\lambda$  and the Bragg angle corresponding to it  $\theta_B$ , then because of the smallness of  $\lambda - \lambda_0$ , it is possible to expand  $\theta_B$  in a power series, keeping only two terms, thus:

$$\theta_B = \theta_0 + (\lambda - \lambda_0) \frac{\partial \theta_0}{\partial \lambda} \quad (11)$$

where

$$\frac{\partial \theta_0}{\partial \lambda} = \frac{1}{2d \cos \theta_0} = \frac{\tan \theta_0}{\lambda_0}$$

obtained by differentiating the Bragg equation  $\lambda_0 = 2d \sin \theta_0$ .

The intensity reflected by the crystal is a function of the difference between the grazing angle and the Bragg angle.

Subtracting (11) from (9), we get

$$\theta - \theta_B = t - \gamma - (\lambda - \lambda_0) \frac{\partial \theta_0}{\partial \lambda} \quad (12)$$

or upon replacing  $\gamma$  and  $\frac{\partial \theta_0}{\partial \lambda}$  by their equivalent expressions

$$\theta - \theta_B = t - \frac{z^2 \tan \theta_0}{8 \sin^2(\theta_0 - \alpha)} - \frac{\lambda - \lambda_0}{\lambda_0} \tan \theta_0 \quad (12a)$$



The intensity of the diffraction pattern of the crystal can be written as  $F_{\frac{\sigma}{\pi}} = F_{\frac{\sigma}{\pi}} (\theta - \theta_B)^*$  and its shape in the case of no absorption is given by Compton and Allison<sup>(8)</sup>, Fig. VI-7. The full width at half maximum of the diffraction pattern, in the case of the (310) planes of quartz, is approximately 2 seconds of arc. This means that for  $\theta - \theta_B > 1''$  only a small fraction of the intensity of wavelength  $\lambda$  is reflected. In the preceding calculations angles that are less than the diffraction pattern width were neglected. The same will be done in the calculations that follow. It is justifiable to drop terms of order  $2''$  or less since such terms do not reveal anything of interest regarding the focusing properties of the laminae.

Let the direction cosines of the reflected ray be k, m, n. These must satisfy the following three equations.

$$k^2 + m^2 + n^2 = 1 \quad (13)$$

$$k \cos(\theta_0 + \eta) + m \sin(\theta_0 + \eta) = -\sin \theta \quad (14)$$

$$\begin{vmatrix} k & m & n \\ \cos(\theta_0 + \eta) & \sin(\theta_0 + \eta) & 0 \\ \frac{\sin(2\theta_0 + \eta + t)}{a} & \frac{-\cos(2\theta_0 + \eta + t)}{a} & \frac{b}{a} \end{vmatrix} = 0$$

where eq. (14) insures that the reflected ray makes an angle

---

\*  $\sigma$  and  $\pi$  refer to the two components of polarization; the former has its electric vector in the plane of incidence and the latter at right angles to it. In the case of no absorption  $F_{\frac{\sigma}{\pi}} = I_{\frac{\sigma}{\pi}}^D$  where  $I_{\frac{\sigma}{\pi}}^D$  are the Darwin functions for the two types of polarization, given in Compton and Allison p. 391.

$\pi/2 - \theta$  with the normal to the crystal planes,  $\overline{PB}$ , and the determinant insures that it be coplanar with  $\overline{PB}$  and  $\overline{SP}$ . The determinant reduces to:

$$k \sin(\theta_0 + \eta) - m \cos(\theta_0 + \eta) = \frac{n}{b} \cos(\theta_0 + t) \quad (15)$$

Multiplying (14) and (15) first by  $\cos(\theta_0 + \eta)$  and  $\sin(\theta_0 + \eta)$  respectively and adding, and then by  $\sin(\theta_0 + \eta)$  and  $-\cos(\theta_0 + \eta)$  respectively and adding, one obtains

$$k = (n/b) \cos(\theta_0 + t) \sin(\theta_0 + \eta) - \sin\theta \cos(\theta_0 + \eta) \quad (16)$$

$$m = -(n/b) \cos(\theta_0 + t) \cos(\theta_0 + \eta) - \sin\theta \sin(\theta_0 + \eta) \quad (17)$$

which, when substituted in (13), gives

$$n^2 = \frac{b^2 \cos^2 \theta}{b^2 + \cos^2(\theta_0 + t)}$$

From (7)

$$\cos^2 \theta = \frac{b^2 + \cos^2(\theta_0 + t)}{a^2} \quad (18)$$

Substituting (18) in the expression for  $n^2$  and taking the square root, we obtain

$$n = b/a \quad (19)$$

Finally upon substitution of (7) and (19) in (16) and (17), the direction cosines  $k$ ,  $m$ ,  $n$ , are reduced to their simplest form

$$\begin{aligned} k &= (1/a) \sin(\eta - t) \\ m &= -(1/a) \cos(\eta - t) \\ n &= b/a \end{aligned} \quad (20)$$

Note that the direction cosines in the  $z$  direction for both the incident and reflected ray are the same. This is obvious if one thinks of the crystal as being a plane mirror in the  $z$  direction.

Assuming the second crystal is removed momentarily, then the projection of the ray, represented by  $k, m, n$ , on the focal plane is the line  $\overline{PF}$  where  $F$  is displaced from  $F_0$  by an angle  $2t$ , just as  $S$  is with respect to  $S_0$ . Since the position of the point  $F$  depends on the variables  $t$  and  $t_z$  only, and not on  $\eta$ , then all rays from  $S$  will focus in a straight vertical line passing through  $F$ . Furthermore, since no approximations were involved in deriving the expressions for  $k, m, n$ , the focusing occurs without any aberrations, however, the line through  $F$  contains all wavelengths in the neighborhood of  $\lambda_0$ . To see this, we must consider eq. (12a) and note that reflections occur only when  $\theta - \theta_B = 0$ , where the equality holds to within the diffraction pattern width. Thus, if  $S$  and hence  $t$  is fixed, then  $\lambda$  is a function of  $z$ , with  $\lambda$  decreasing with increasing  $|z|$ .

Since to each point  $S$  there corresponds a point  $F$ , this implies that given a sufficiently wide spectral band of wavelengths the width of the line focus could be essentially the width of the target, approximately 1 mm. However, neglecting the continuous x-ray spectrum, the range of wavelengths permitted to go through is approximately 3 times the full width at half maximum of the  $K\alpha_1$  line, so that the line focus appears to be about 1/3 mm. wide.

If instead of keeping  $t$  fixed, as was done above,  $\lambda$  is kept fixed, then (12a) shows the dependence of  $t$  on  $z$ . The dependence is such that "lines" of equal  $\lambda$  are parabolas whose

branches extend in the positive  $t$  direction, i.e., toward the longer wavelengths.

In order to be able to trace the ray any further, it is necessary to describe it in the coordinate system of the second crystal  $x', y', z'$ . The latter axes are oriented in the same manner with respect to the second crystal as  $x, y, z$  are with respect to the first.

The relation between the two coordinate systems is given by

$$\begin{aligned}x' &= \cos(\theta_0 + \alpha) - z \\y' &= 2 \sin(\theta_0 - \alpha) - y \\z' &= \cos(\theta_0' + \alpha) - x\end{aligned}\tag{21}$$

where  $\cos(\theta_0 + \alpha) = \overline{CC_P} = \overline{C'C'_P}$  and  $2 \sin(\theta_0 - \alpha) = \overline{C_P C'_P}$  as seen from Fig. 5.

Let the direction cosines of the reflected ray in the primed coordinate system be  $k', m', n'$ , and in view of the above transformations, and eqs. (20)

$$\begin{aligned}k' &= -n = -b/a \\m' &= -m = \cos(\eta - t)/a \\n' &= -k = -\sin(\eta - t)/a\end{aligned}\tag{22}$$

Let the coordinates of the point  $P$  in the primed system be  $x'_1, y'_1, z'_1$ , then

$$\begin{aligned}x'_1 &= \cos(\theta_0 + \alpha) - z \\y'_1 &= 2 \sin(\theta_0 - \alpha) - \sin \phi \\z'_1 &= \cos(\theta_0 + \alpha) - \cos \phi\end{aligned}$$

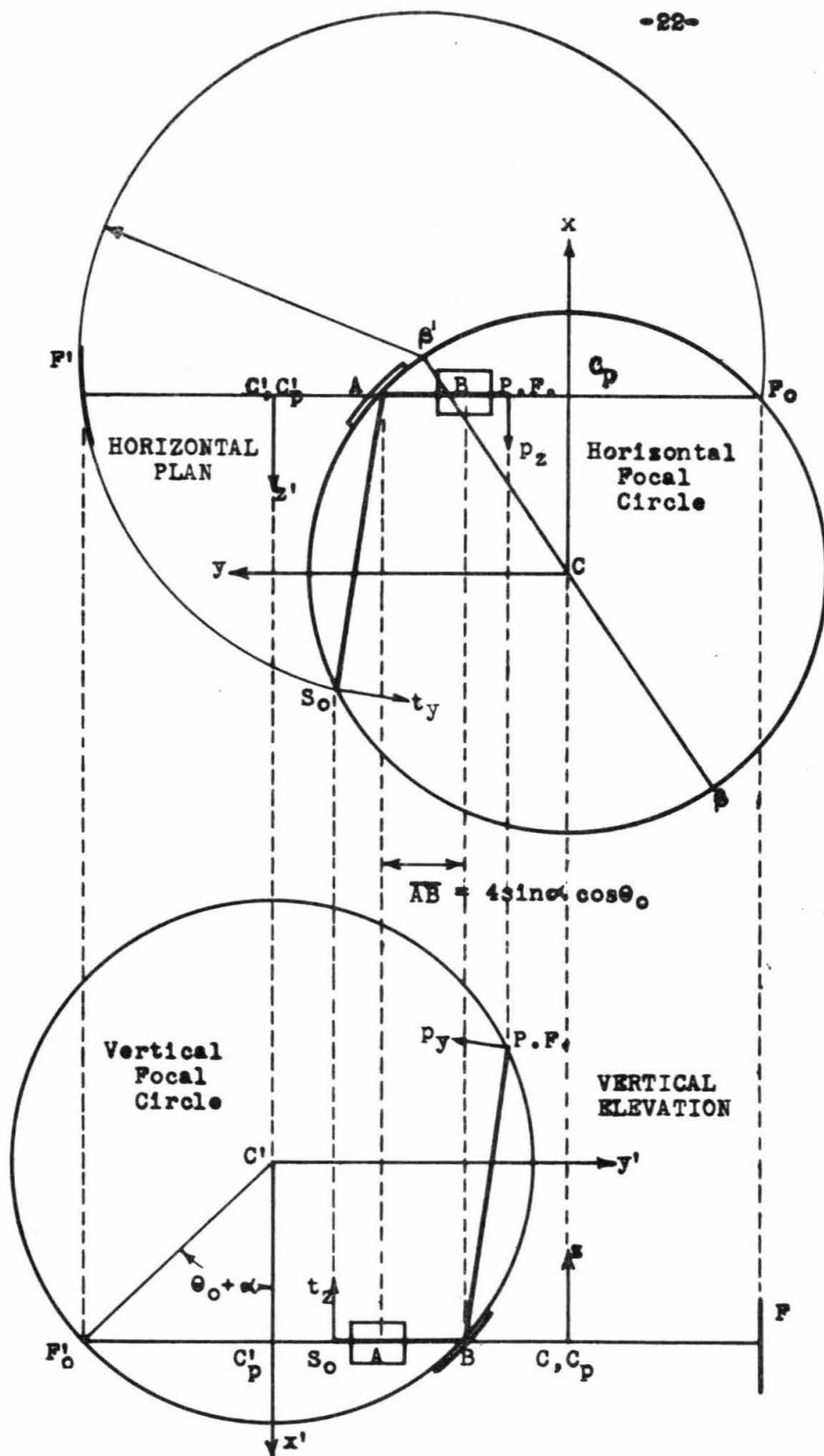


Fig. 5 - Horizontal and vertical focal circles shown together with their respective coordinate axes.

Also let the coordinates of the point P', where the ray strikes the second crystal, be x', y', z' which in terms of cylindrical coordinates are given by

$$x' = \cos \phi'$$

$$y' = \sin \phi'$$

$$z' = z'$$

so that

$$\frac{\cos \phi' - \cos(\theta_0 + \alpha) + z}{n} = \frac{\sin \phi' - 2\sin(\theta_0 - \alpha) + \sin \phi}{m} = \frac{z' - \cos(\theta_0 + \alpha) + \cos \phi}{k}$$

or

$$\cos \phi' = \cos(\theta_0 + \alpha) - z + (n/m) [\sin \phi' - 2\sin(\theta_0 - \alpha) + \sin \phi] \quad (23)$$

$$z' = \cos(\theta_0 + \alpha) - \cos \phi + (k/m) [\sin \phi' - 2\sin(\theta_0 - \alpha) + \sin \phi] \quad (24)$$

It is necessary to solve eq. (23) for  $\phi'$ , or for  $\eta'$ , where by analogy to eq. (1), we let,

$$2\eta' = \phi' - \theta_0 - \alpha \quad (25)$$

where  $2\eta'$  is the central angle  $\widehat{BC'P'}$  so that,

$$\cos \phi' \approx (1 - 2\eta'^2) \cos(\theta_0 + \alpha) - 2\eta' \sin(\theta_0 + \alpha) \quad (26)$$

$$\sin \phi' \approx \sin(\theta_0 + \alpha) + 2\eta' \cos(\theta_0 + \alpha)$$

Similarly from (1)

$$\cos \phi \approx (1 - 2\eta^2) \cos(\theta_0 + \alpha) - 2\eta \sin(\theta_0 + \alpha) \quad (27)$$

$$\sin \phi \approx \sin(\theta_0 + \alpha) + 2\eta \cos(\theta_0 + \alpha)$$

From (20)

$$\frac{n}{m} = \frac{b}{-\cos(\eta - t)} \approx -b \quad (28)$$

where from (5)

$$b \approx \frac{z [1 + \eta \cot(\theta_0 - \alpha)] - t_z}{2 \sin(\theta_0 - \alpha)} \quad (29)$$



Substituting eqs. (26 to 29) in (23), and neglecting terms of order higher than  $z^2$ ,  $\eta^2$  or  $z\eta$ , as was done above, a quadratic equation in  $\eta'$  is obtained, which is as follows:

$$\eta'^2 \cot(\theta_0 + \alpha) + \eta' = \frac{z}{2 \sin(\theta_0 - \alpha)} \left[ 1 + \frac{z \cot(\theta_0 + \alpha)}{2 \sin(\theta_0 - \alpha)} + \eta \cot(\theta_0 - \alpha) - \frac{2\eta \sin \alpha \sin \theta_0}{\sin(\theta_0 + \alpha)} \right] - \frac{tz \sin \alpha \cos \theta_0}{\sin(\theta_0 - \alpha) \sin(\theta_0 + \alpha)}$$

By substitution of  $|z| \leq 1/60$ ,  $|\eta| \leq 1/80$ , and  $\alpha \approx 1/30$  radians, it is found that the next to the last term in the preceding equation is of the order of  $2''$  and hence may be neglected.

The quadratic equation is of the form

$$A \eta'^2 + \eta' = B$$

whose solution is

$$\eta' = \frac{-1 + (1 + 4AB)^{1/2}}{2A} \approx B - AB^2$$

Applying the above equation to (30), the following result is obtained,

$$\eta' = \frac{z}{2 \sin(\theta_0 - \alpha)} \left[ 1 + \eta \cot(\theta_0 - \alpha) \right] - \frac{2t' \sin \alpha \cos \theta_0}{\sin(\theta_0 - \alpha)} \quad (31)$$

where

$$t' = t_z / 2 \sin(\theta_0 + \alpha)$$

and plays the same role in the primed coordinate system as  $t$  does in the unprimed system,  $|t'| \leq 1/1500$  radians.

To solve for  $z'$ , we substitute  $k/m = -\tan(\eta - t)$  together with eqs. (26), (27) and (31) into (24), which after simplification becomes

$$z' = 2\eta \sin(\theta_0 - \alpha) - \frac{z\eta \cos(\theta_0 + \alpha)}{\sin(\theta_0 - \alpha)} + 4t \sin \alpha \cos \theta_0 \quad (32)$$

The direction cosines of the normal to the crystal planes at  $P'$  are given by (6), with  $\eta'$  replacing  $\eta$ . They are:

$$\cos(\theta_0 + \eta'), \sin(\theta_0 + \eta'), 0$$

The direction of the ray incident on the second crystal  $k', m', n'$  are given in terms of  $b, \eta, t$  by eqs. (22).

Let the angle between the above two directions be  $\pi/2 - \theta'$  where  $\theta'$  is the grazing angle. Therefore,

$$\begin{aligned} \cos\left(\frac{\pi}{2} - \theta'\right) &= \sin\theta' = k' \cos(\theta_0 + \eta') + m' \sin(\theta_0 + \eta') \\ &\approx -b \cos(\theta_0 + \eta') + (1 - b^2/2) \cos(\eta - t) \sin(\theta_0 + \eta') \\ &\approx -\left[b + \frac{(\eta - t)^2}{2} \tan(\theta_0 + \eta')\right] \cos(\theta_0 + \eta') \\ &\quad + (1 - b^2/2) \sin(\theta_0 + \eta') \end{aligned} \quad (34)$$

where eqs. (5), (8) and (22) were used in the second step, and in the last step  $\cos(\eta - t)$  was expanded into  $1 - (\eta - t)^2/2$ .

Let

$$\gamma' = b + \frac{(\eta - t)^2}{2} \tan(\theta_0 + \eta') \quad (35)$$

Since  $\gamma'$  is a small quantity, it is permissible to rewrite eq. (34), thus:

$$\begin{aligned} \sin\theta' &\approx -\sin \gamma' \cos(\theta_0 + \eta') + \cos \gamma' \sin(\theta_0 + \eta') \\ &= \sin(\theta_0 + \eta' - \gamma') \end{aligned}$$

from which it is easily seen that

$$\theta' = \theta_0 + \eta' - \gamma'$$

With the help of (29), eq. (35) can be simplified by dropping higher order terms, namely those involving  $t$ . Thus,

$$\gamma' = \frac{z}{2 \sin(\theta_0 - \alpha)} \left[ 1 + \cot(\theta_0 - \alpha) \right] + \frac{\eta^2}{2} \tan \theta_0 - \frac{t' \sin(\theta_0 + \alpha)}{\sin(\theta_0 - \alpha)}.$$

Combining the above with (31), results in

$$\theta' = \theta_0 + t' - \frac{\eta^2}{2} \tan \theta_0 \quad (36)$$

As in the case of the first reflection, we are interested in the difference between the grazing angle  $\theta'$ , and the Bragg angle  $\theta_B$  for the wavelength  $\lambda$ . Making use of (11), the final result is,

$$\theta' - \theta_B = t' - \frac{\eta^2}{2} \tan \theta_0 - \frac{\lambda - \lambda_0}{\lambda_0} \tan \theta_0 \quad (37)$$

The above equation can now be compared with (12a), which is,

$$\theta - \theta_B = t - \frac{z^2 \tan \theta_0}{8 \sin^2 (\theta_0 - \alpha)} - \frac{\lambda - \lambda_0}{\lambda_0} \tan \theta_0 \quad (38)$$

The conditions that must be imposed on  $(\theta - \theta_B)$  and  $(\theta' - \theta_B)$  in order that a ray of wavelength  $\lambda$  be reflected by both crystals are,

$$\theta - \theta_B = \theta' - \theta_B = 0 \quad (39)$$

where the equality is to be understood to hold to within the diffraction width, namely  $2''$  of arc. This, incidentally, is another justification for dropping small order terms.

In view of the above requirements, eqs. (37) and (38) show some interesting properties of the crystals. For purposes of computation it is better to rewrite the above equations in

numerical form by substituting  $\theta_0 = 40.7^\circ$  and  $\alpha = 2.1^\circ$ ,

$$\theta' - \theta_B = t' - 0.430 \eta^2 - 0.860(\lambda - \lambda_0)/\lambda_0 \quad (40)$$

$$\theta - \theta_B = t - 0.276z^2 - 0.860(\lambda - \lambda_0)/\lambda_0 \quad (41)$$

Subtracting (37) from (38), gives the dependence of  $\eta$  and  $z$  on  $t$  and  $t'$  alone,

$$(\eta^2/2)\tan\theta_0 - z^2\tan\theta_0/8 \sin^2(\theta_0 - \alpha) = t' - t \quad (42)$$

or

$$0.430 \eta^2 - 0.276z^2 = t' - t \quad (42a)$$

If the target of the x-ray tube is so small that  $t$  and  $t'$  can be neglected, or if  $t' - t = 0$  in the above equation, then the area of the first crystal which is contributing to the point focus degenerates into two lines whose widths ( $\approx 0.01$  mm.) correspond to the diffraction pattern widths and whose equations are

$$z = \pm 1.25 \eta \quad (43)$$

along which the wavelength decreases with increasing  $|z|$ , as is given by eq. (41). By definition,  $2\eta$  is the angle measured at the center  $C$  of the focal circle from the center of the crystal to the arbitrary point  $P$ , so that  $z$  and  $2\eta$  may be considered as the coordinate axes of the crystal lamina with the origin at its center. If the above two lines are projected on a plane perpendicular to the incident (or once reflected) beam, then they become orthogonal and are inclined at  $45^\circ$  with the horizontal plane. Since the reflecting portions of the crystal laminae were designed to admit a square beam,  $3/4" \times 3/4"$ , it is obvious that the above lines connect the opposite corners of the laminae. When  $t' - t$  is not negligible, eq. (42) represents

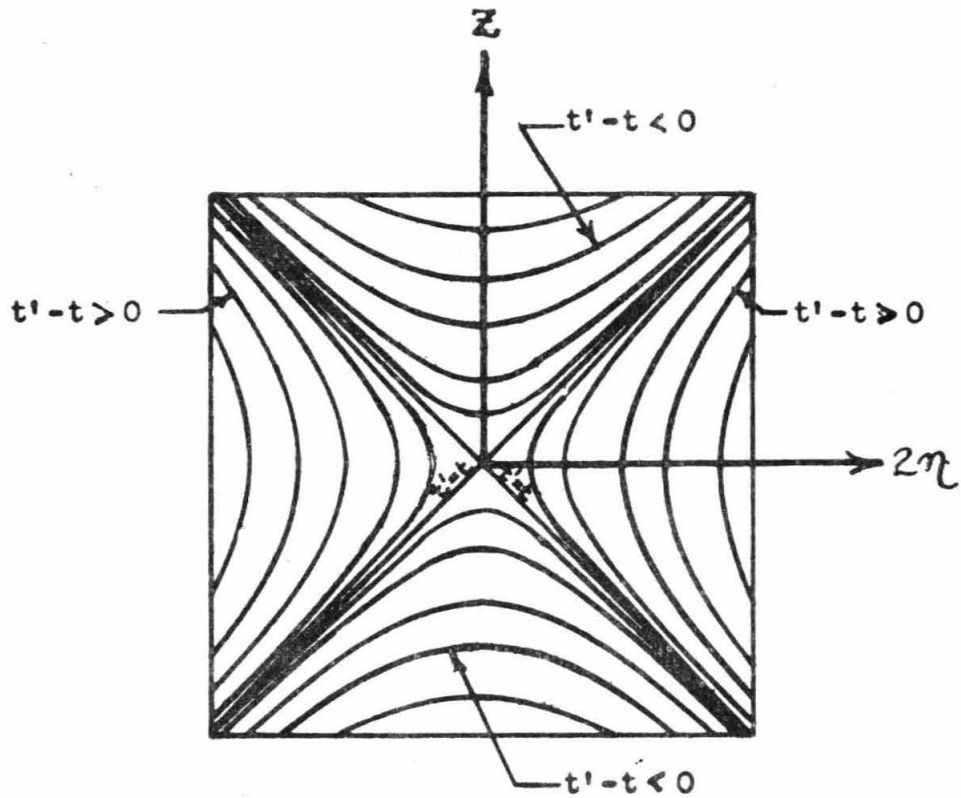


Fig. 6 - Cross-section of beam ( $3/4'' \times 3/4''$ ) midway between the two crystals. Hyperbolas represent cross-section of rays emanating from points in the target for which  $t' - t$  is a constant.

two families of hyperbolas whose asymptotes are eqs. (43). The upper and lower hyperbolas correspond to negative values of  $t' - t$ . When  $|t' - t|$  takes on all values from zero to some finite value, eq. (42a) gives the four limiting hyperbolas within which the first crystal lamina is illuminated by x-rays. Eq. (42a) also shows that in order that the whole lamina be illuminated  $t' - t$  must satisfy the following inequality,

$$-0.042 \leq t' - t \leq 0.042 \text{ mm.}$$

This inequality shows that the useful target area is a strip inclined at approximately  $45^\circ$  with the horizontal, and extending from the upper left hand corner to the lower right hand corner of the target. It must be remembered that the values  $t$  and  $t'$  do not represent true distances from the center of the target as seen from the center of crystal A, but must be multiplied by  $2 \sin(\theta_0 + \alpha)$  and  $2 \sin(\theta_0 - \alpha)$  respectively, to give  $t_z$  and  $t_y$ . Thus,  $t' - t = t_z/2 \sin(\theta_0 + \alpha) - t_y/2 \sin(\theta_0 - \alpha)$  so that,

$$-0.06 \leq t_z - 1.09 t_y \leq 0.06 \text{ mm.} \quad (43.1)$$

and the actual inclination of the strip is  $47 \frac{1}{2}^\circ$  with the horizontal plane.

The limits on  $t$  and  $t'$  individually are given by (40) and (41) in which the maximum value of  $|\lambda - \lambda_0|$  can be taken arbitrarily as 0.30 X.U., i.e. the half width at half maximum of the  $K\alpha_1$  line. Hence,

$$-0.10 \leq t \leq 0.14 \text{ mm.} \quad \text{or} \quad -0.13 \leq t_y \leq 0.17 \text{ mm.}$$

$$-0.10 \leq t' \leq 0.15 \text{ mm.} \quad \text{or} \quad -0.14 \leq t_z \leq 0.20 \text{ mm.}$$



It is seen that the useful target area is a diagonal strip about 0.09 mm. wide enclosed in a rectangular area 0.30 mm. x 0.34 mm., the sides of which are not symmetrically disposed about the center of the target ( $t = t' = 0$ ), see Fig. 8a, p. 38. Note that the width of the strip is independent of the wavelength, except through the factor  $\theta_0$ , as given by eq. (42).

It is known that the curves representing the intensity versus wavelength of x-ray spectral lines are "witches".\* Thus, it is obvious that the area under that part of the witch (out to half-max.), which is being reflected above, is half the total area. If the full target, 1 mm. x 1 mm., is used instead, then it is not hard to show that about 83% of the area under the witch is being reflected. The useful area of the target, however, is still a diagonal strip of the same width, namely 0.09 mm. This width is independent of wavelength admitted, but depends on the size of the crystal laminae only.

Returning our attention to the ray, which was found to strike the second crystal with a grazing angle  $\theta'$  at the point  $P'$  whose coordinates are  $(1, \theta_0 + \alpha + 2\eta', z')$ , we now consider the twice reflected ray and follow it through to the point focus.

Let the direction cosines of the twice reflected ray be  $k''$ ,  $m''$ ,  $n''$ , then the conditions that they must satisfy are similar to those given by (13), (14) and the determinant, with

---

\*The standard equation of a "witch" at the origin is

$$\frac{A}{1 + x^2/w^2}.$$

$\eta'$  replacing  $\eta$  and  $k'$ ,  $m'$ ,  $n'$  replacing the last row in the determinant.

$$k''^2 + m''^2 + n''^2 = 1 \quad (44)$$

$$k'' \cos(\theta_0 + \eta') + m'' \sin(\theta_0 + \eta') = -\sin \theta' \quad (45)$$

$$\begin{vmatrix} k'' & m'' & n'' \\ \cos(\theta_0 + \eta') & \sin(\theta_0 + \eta') & 0 \\ k' & m' & n' \end{vmatrix} = 0$$

As in the case of the first reflection, the direction cosines in the  $z'$  direction of the incident and reflected rays are equal, i.e.  $n'' = n'$ , so that with the help of (22) we have:

$$n'' = -(1/a) \sin(\eta - t) \quad (46)$$

Subtracting the last row of the determinant from the first and expanding, we get

$$(k'' - k') \sin(\theta_0 + \eta') = (m'' - m') \cos(\theta_0 + \eta')$$

Eliminating  $\sin \theta'$  from (45) by means of (34), we get

$$(k'' + k') \cos(\theta_0 + \eta') = -(m'' + m') \sin(\theta_0 + \eta')$$

Solving the last two equations for  $k''$  and  $m''$ , we obtain

$$k'' = -k' \cos 2(\theta_0 + \eta') - m' \sin 2(\theta_0 + \eta') \quad (47)$$

$$m'' = -k' \sin 2(\theta_0 + \eta') + m' \cos 2(\theta_0 + \eta') \quad (48)$$

which are left in the present form for convenience.

In the ideal case the position of the point focus, relative to the primed coordinate system, must be the same as that of the center of the target relative to the unprimed system. The coordinates of the center of the target, point  $S_0$ , are  $(1, 3\theta_0 - \alpha, 0)$  and hence the coordinates of the point focus must be the same. The general ray that we are dealing with,

however, may not pass exactly through this point, but may pass close by, say through  $(1, 3\theta_0 - \alpha + 2p, p_z)$  where  $p$  and  $p_z$  are small quantities. If we let

$$p_y = 2p \sin(\theta_0 - \alpha),$$

then  $p_y$  and  $p_z$  represent the coordinates of the general ray in a plane perpendicular to the central ray and passing through the point where it touches the focal circle, the coordinates of the central ray being  $(0,0)$ . See Fig. 5.

The equation of the twice reflected ray can now be written as:

$$\frac{\cos(3\theta_0 - \alpha + 2p) - \cos\phi'}{k''} = \frac{\sin(3\theta_0 - \alpha + 2p) - \sin\phi'}{m''} = \frac{p_z - z'}{n''} \quad (49)$$

Substituting (47) and (48) in the first two terms above, and simplifying without making any approximations, we get

$$\tan(p - \eta') = k'/m' \quad (50)$$

where use has been made of  $\phi' = \theta_0 + \alpha + 2\eta'$ . From eqs. (22),  $k'/m' = n/m = -b/\cos(\eta - t) \approx -b$ , and since  $\tan(p - \eta') \approx p - \eta'$ , eq. (50) becomes, with the help of (31) and (29),

$$p = \eta' - b = t' \quad (51)$$

or

$$p_y = t_z \sin(\theta_0 - \alpha) / \sin(\theta_0 + \alpha) \quad (52)$$

To find the coordinates of the ray in the other direction, i.e.  $p_z$ , we consider again eq. (49),

$$p_z = z' + (n''/k'') [\cos(3\theta_0 - \alpha + 2p) - \cos\phi']$$

where  $z'$  is given by (32) and  $n''/k''$  is given by (46), (47) and (22). The resulting equation is,

$$p_z \approx 2t \sin(\theta_0 + \alpha) = t_y \sin(\theta_0 + \alpha) / \sin(\theta_0 - \alpha) \quad (53)$$

Subtracting (53) from (52), we get

$$p_y \sin(\theta_0 + \alpha) / \sin(\theta_0 - \alpha) - p_z = t_z - t_y \sin(\theta_0 + \alpha) / \sin(\theta_0 - \alpha) \quad (54)$$

or numerically

$$1.09p_y - p_z = t_z - 1.09t_y \quad (54a)$$

which in view of the inequality (43.1) gives

$$-0.06 \leq p_z - 1.09p_y \leq 0.06 \text{ mm.} \quad (55)$$

Eqs. (52) and (53) together with the inequalities imposed on  $t_z$  and  $t_y$ , for transmission of half the area under the  $K\alpha_1$  line, give

$$-0.13 \leq p_y \leq 0.18 \text{ mm.} \quad (56)$$

$$-0.14 \leq p_z \leq 0.18 \text{ mm.}$$

The three inequalities above show that the focal spot in the final "point" image is a strip of width 0.09 mm., inclined at  $47 \frac{1}{2}^\circ$  with the vertical focal plane. This strip is enclosed in a rectangular area 0.31 mm. x 0.32 mm. See Fig. 8a, p. 38.

We can now come to the following conclusion. The focal spot is the image of the useful portion of the target, namely, a diagonal strip 0.09 mm. wide. By varying the target aperture, it is possible to change the shape and size of the focal spot. Although the focal spot is the image of the target, the resolution of the instrument is determined by its geometry. In other words, no matter what size target is used, only a diagonal strip is useful in forming a focal spot. The length of the strip is determined by the width of the  $K\alpha_1$  line; beyond that only the continuous radiation is reflected. If the strip

is sufficiently long, i.e. if the target extends sufficiently along the length of the strip, about 1.8 mm., the  $K\alpha_2$  line may be reflected. The latter case is shown by means of dotted lines in Fig. 8a, p. 38. This, however, is not desirable since the focal spot will then appear as two elongated dots. Any target area outside the diagonal strip is useless and only contributes to the incoherent scattering which adds to the general background in the neighborhood of the focal spot. If a tube whose target area is 1 mm. x 1 mm. is used, the focal spot is approximately 1.4 mm. x 0.09 mm.

Before concluding this section, it must be pointed out that the intensity reflected by the two crystals is a complicated expression involving the integral  $F_{\sigma}(\theta - \theta_B) F_{\pi}(\theta' - \theta_B)$  where the arguments of the functions  $F$  are given by eqs. (40) and (41). The intensity problem will be discussed in more detail later.

#### ABERRATIONS

So far, only the ideal focusing system has been considered, that is, the case where the crystals have been cylindrically ground prior to bending. In practice, however, it is found to be a rather difficult task to grind crystals cylindrically with any degree of accuracy. Instead, the crystals are ground flat and then bent to twice the radius of the focal circle in order to obtain the proper curvature for the crystal planes. Thus, the concave crystal face does not everywhere hug the focal circle but is only tangent to it at the center of the crystal.

The use of initially flat crystals and their effect on the focusing qualities of single crystal monochromators has been discussed extensively by Johann<sup>(10)</sup>. Herein, we shall designate this as the approximate or inexact focusing system.

To find what happens to the focal spot when two such approximate focusing crystals are used in the point focusing arrangement, it is necessary to retrace a general ray through the system. Since it is neither instructive nor interesting to repeat the calculations in detail, only the more important results will be presented.

Fig. 7 shows a flat crystal which has been bent to a radius equal to twice the focal radius. Point P is again designated as a general point on the crystal lamina, however, it is no longer on the focal circle.  $C_f$  is the center of curvature of the crystal face and  $\eta$  is measured about the point  $C_f$ .

The difference between the grazing angle at the first crystal and the Bragg angle for the wavelength  $\lambda$  is given by

$$\theta - \theta_B = t - 0.276z^2 - 0.860(\lambda - \lambda_0)/\lambda_0 + 0.628\eta^2 \quad (57)$$

At the second crystal it is found that

$$\theta' - \theta_B = t' - 0.430\eta^2 - 0.860(\lambda - \lambda_0)/\lambda_0 + 0.350z^2 \quad (58)$$

Comparing these equations with (41) and (40), it is seen that they differ only in the last term.

In order that a ray be reflected by both crystals, the two previous equations must be equated to zero as has been done before. Subtracting (57) from (58), we get an equation similar to (42),

$$1.058\eta^2 - 0.626z^2 = t' - t \quad (59)$$



representing two families of hyperbolas whose asymptotes are

$$z = \pm 1.30 \eta$$

It is thus seen that, in passing from the ideal to the approximate focusing system, the reflecting features of the two crystals are not altered appreciably, although in this latter scheme the inequalities to be imposed on  $t$ ,  $t'$  and  $t' - t$  are given by (57), (58) and (59).

$$\begin{aligned} -0.16 \leq t \leq 0.14 \text{ mm.} & \quad \text{or} \quad -0.20 \leq t_y \leq 0.17 \text{ mm.} \\ -0.16 \leq t' \leq 0.15 \text{ mm.} & \quad \text{or} \quad -0.22 \leq t_z \leq 0.20 \text{ mm.} \quad (60) \\ -0.10 \leq t' - t \leq 0.10 \text{ mm.} & \quad \text{or} \quad -0.14 \leq t_z - 1.09 t_y \leq 0.14 \text{ mm.} \end{aligned}$$

The useful target area is again a diagonal strip, but with width 0.19 mm. enclosed in a rectangular area 0.37 mm. x 0.42 mm., as is shown in Fig. 8b, p. 38. Although the length of the strip is about the same as in the ideal case, the width is doubled.

The focal spot is again the image of the useful target area, namely, a strip 0.19 mm. wide inclined at  $47 \frac{1}{2}^\circ$  with the vertical focal plane (focal plane of the second crystal). The extension of the focal spot in the  $y$  and  $z$  directions (for transmission of half the area under the  $K\alpha_1$  line) is given by,

$$\begin{aligned} -0.20 \leq p_y \leq 0.16 \text{ mm.} \\ -0.22 \leq p_z \leq 0.19 \text{ mm.} \end{aligned} \quad (61)$$

Hence, the strip is enclosed in a rectangular area 0.36 mm. x 0.41 mm. The whole target (1 mm x 1 mm.) will transmit about 81% of the area under the  $K\alpha_1$  line. The focal spot will then be approximately 0.19 mm. x 1.41 mm. It is interesting to note that an



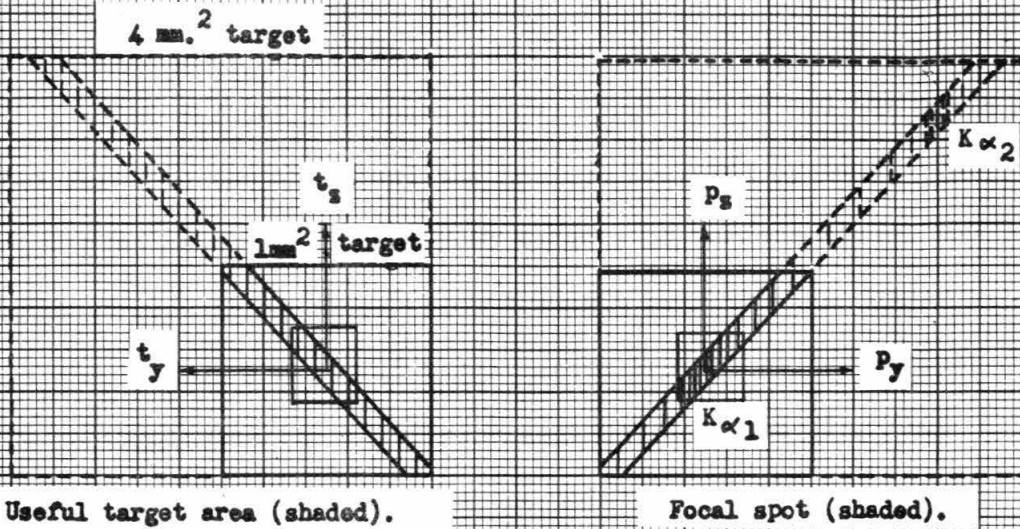


Fig. 8a - Useful target area and focal spot in the exact focusing system.

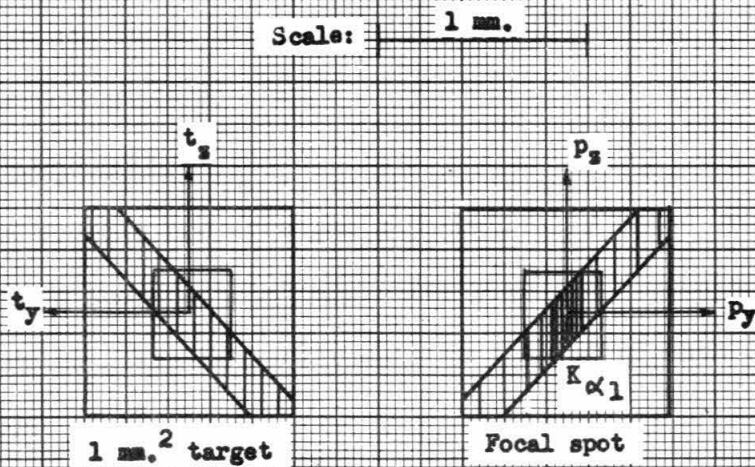


Fig. 8b - Useful target area and focal spot in the inexact focusing system.

actual photograph of the focal spot reveals its size to be 0.196 mm. x 1.40 mm. when measured under a traveling microscope, see Plate 9, p. 70.

Fig. 8 shows the useful x-ray target areas and the point foci for both the exact and the inexact focusing systems. The rectangles on the left represent targets of various sizes, the smallest ones being those which transmit half the area under the  $K\alpha_1$  line. The useful target areas are the shaded strips. The rectangles on the right represent the corresponding (but not equal) areas which enclose the point foci. The latter are the shaded strips with the darker shade representing roughly the more intense portion of the foci. The directions of the coordinates  $t_y$ ,  $t_z$ ,  $p_y$  and  $p_z$  are shown in Fig. 5, p. 22.

This concludes the geometrical consideration of the point focusing monochromator.

#### TABLE OF NOMENCLATURE

The symbols are arranged alphabetically under two headings, Greek Letters and Roman Letters. The page and figure where they first occur is given in parenthesis at the end of each descriptive paragraph.

##### GREEK LETTERS

- $\alpha$  The dihedral angle between the crystal plane and its face at the center of the latter (or between their normals). (P. 9 and Fig. 2).
- $\mathfrak{B}$  The generator of the focal cylinder about which the crystal planes are concentric. (P. 6 and Fig. 2).

- $\beta'$  The point in the focal plane diametrically opposite from  $\beta$ . (P. 8 and Fig. 2).
- $\gamma$  The small angle, referred to the horizontal focal plane, by which the grazing angle of incidence ( $\theta$ ) differs from the Bragg angle ( $\theta_0$ ), due to the vertical divergence of the incident ray. (The vertical divergence angle is the angle between the incident ray and the focal plane.) (P. 16).
- $\gamma'$  Small angle referred to the vertical focal plane and defined by Eq. (35). (P. 25).
- $\eta$  Half the central angle measured about the center of the horizontal focal circle (C), and in its plane, from the center of the first crystal (A) to the arbitrary point P. (P. 14 and Fig. 4). (Note: See also Fig. 7 for definition of  $\eta$  in the inexact focusing system.)
- $\eta'$  Same as  $\eta$  but referred to the vertical focal circle, the center of the second crystal (B) and the point P'. (P. 23).
- $\theta$  The grazing angle or the angle between the ray and the crystal planes of the first crystal at the arbitrary point P. (pp. 8, 15 and Fig. 2).
- $\theta'$  The grazing angle or the angle between the (once reflected) ray and the crystal planes of the second crystal at the point P'. (P. 23).
- $\theta_0$  The Bragg angle for the wavelength  $\lambda_0$  at the center of the  $K\alpha_1$  line. (P. 12 and Fig. 4).
- $\theta_B$  The Bragg angle for any wavelength  $\lambda$  in the neighborhood of  $\lambda_0$ . (P. 15).
- $\lambda_0$  Wavelength at the center of the  $K\alpha_1$  line. (P. 11).
- $\lambda$  Wavelength at points other than the center of the  $K\alpha_1$  line. (P. 17).
- $\phi$  One of three cylindrical coordinates of the point P, the three being  $l$ ,  $\phi$  and  $z$ , with the latter perpendicular to the horizontal focal circle. (P. 11 and Fig. 4).

- $\emptyset'$  Same as  $\emptyset$  but for point  $P'$  and the vertical focal circle. (P. 23).

ROMAN LETTERS

- A Geometrical center of 1st crystal (or crystal with horizontal focal circle.) (P. 8 and Fig. 2).
- $a_0$  Abbreviation for expression on p. 14.
- a Abbreviation for expression on p. 15.
- B Geometrical center of 2nd crystal (or crystal with vertical focal circle). (P. 10 and Fig. 3).
- $b_0$  Abbreviation for expression on p. 14.
- b Abbreviation for expression on p. 15.
- C Center of horizontal focal circle. (P. 6 and Fig. 2).
- $C'$  Center of vertical focal circle. (P. 11 and Fig. 3).
- $C_p$  (P. 21 and Fig. 5.)
- $C'_p$  (P. 21 and Fig. 5.)
- $C_f$  Center of curvature of crystal planes in the inexact focusing system. (P. 35 and Fig. 7).
- d Grating spacing of the crystal laminae used in reflection of  $Cu K\alpha_1$  line. (P. 8).
- F Point on vertical line focus at which the reflected ray from the arbitrary point P on the first crystal intersects the focal cylinder, in the absence of the second crystal. (P. 20 and Fig. 4.) (Also, in the qualitative discussion, the line focus of a single crystal, or its midpoint. (P. 6 and Figs. 2, 3) ).
- $F_0$  Center of vertical line focus; also called the "focal point" of the horizontal focal circle. (P. 12 and Fig. 4).

- $F'$  Point on virtual line source (in horizontal plane) from which the arbitrary ray appears to emanate after it is reflected from the 1st crystal. (Fig. 5). (Also, in the qualitative discussion, the virtual line source or its midpoint. (P. 9 and Fig. 3) ).
- $F'_0$  Center of virtual line source (Fig. 5).
- $k, m, n$  Direction cosines of the once reflected ray from the point P on the 1st crystal to the point  $P'$  on the 2nd crystal in the coordinate system of the former. (P. 16).
- $k', m', n'$  Same as above but in the coordinate system of the 2nd crystal. (P. 19).
- $k'', m'', n''$  Direction cosines of the twice reflected ray from the point  $P'$  on the 2nd crystal to the Point Focus. (P. 28).
- P.F. Point Focus. (P. 10 and Fig. 3).
- P Arbitrary point on 1st crystal at which a ray from point S is reflected. (P. 6 and Fig. 2).
- $P'$  Point on second crystal where the once reflected ray (from point P) is again reflected. (P. 23).
- p Small angular displacement measured about the center of the vertical focal circle and in the plane of the latter where the twice reflected ray pierces the focal plane.\* (P. 29).
- $p_y, p_z$  Coordinates in the focal plane\* of the point where the ray pierces the latter.  $p_z$  is in a direction perpendicular to the vertical focal circle and  $p_y$  is at right angles to it. (P. 32 and Fig. 5).
- r Radius of either focal circle. (P. 11).
- $S_0$  Point on horizontal focal circle representing center of x-ray target. (P. 12 and Fig. 4).
- S Point at which the arbitrary incident ray intersects the focal cylinder in the neighborhood of  $S_0$ . (P. 15 and Fig. 4). Also in the qualitative discussion, a point source of x-rays. (P. 6, Fig. 2).

---

\* Plane passing through the point focus at right angles to the central ray.

- $t$  Small angular displacement measured about the center of the horizontal focal circle and in the plane of the latter, where the arbitrary ray emerges from the target. (P. 15, and Fig. 4).
- $t_y, t_z$  Coordinates in a plane at right angles to the central ray through the point  $S_0$  of the arbitrary ray emerging from the target. (P. 15 and Fig. 5).
- $(x, y, z)$  Coordinate system of horizontal focal circle. (P. 11 and Figs. 4, 5).
- $(x', y', z')$  Coordinate system of the vertical focal circle. (P. 21 and Fig. 5).
- $x_1', y_1', z_1'$  Coordinates of point P in the primed coordinate system. (P. 21).

### PART III

#### THE INTENSITY PROBLEM AND SUGGESTED IMPROVEMENTS

The problem of obtaining sufficient intensity is one of the most difficult in connection with the point focusing monochromator and one that has been the cause of a good deal of concern. Losses due to polarization at each reflection are inherent in the design of the instrument and can only be reduced by a decrease in the Bragg angle. (See eq. (64) below.) The latter can be accomplished by one of two means, increasing the grating spacing or decreasing the wavelength, neither of which lends itself to a wide variety of choice.

Assuming the first crystal is illuminated uniformly with x-rays, then the intensity at the point focus is given by,

$$I = (1/2) I_0 C_\sigma C_\pi \int_{-\lambda_{\max}}^{\lambda_{\max}} \int_{-\theta_{\max}}^{\theta_{\max}} \int_{-\theta'_{\max}}^{\theta'_{\max}} J(\lambda - \lambda_0) \left[ F_\sigma(\theta - \theta_B) F_\pi(\theta' - \theta_B) + F_\pi(\theta - \theta_B) F_\sigma(\theta' - \theta_B) \right] d\lambda d\theta d\theta' \quad (62)$$

where it must be remembered that  $(\theta - \theta_B)$  and  $(\theta' - \theta_B)$  are given by eqs. (41) and (40) respectively.

$C_\sigma$  is a factor depending on the thickness of the crystal, its diffraction pattern width, radius of curvature, average extinction coefficient and linear absorption coefficient. According to the dynamical theory of x-ray reflection from crystals, the condition for one hundred percent reflection (in the absence of absorption) occurs when  $\theta - \theta_B$  and  $\theta' - \theta_B$  are less than half the diffraction width. Outside that range

very little reflection occurs. This will be obvious as soon as the function  $F_{\sigma}$  and  $F_{\pi}$  are written down explicitly. When the condition for complete reflection is satisfied at the surface, the incident rays are said to be rapidly "extinguished". In other words, they are reflected before they have a chance to penetrate very far into the crystal. However, when the condition is not satisfied at the surface as is the case with a target of finite size, the rays penetrate into the crystal, suffering attenuation due to absorption until they reach crystal planes for which the condition is satisfied, in which case, they are then rapidly extinguished. This situation is only possible with curved crystals in which the grazing angle changes as the beam penetrates the lattice. The reflecting properties of bent crystals have been the subject of a recent doctoral thesis and have been reported in J. App. Phys. (11)

$J(\lambda - \lambda_0)$  is the spectral distribution function. The functions  $F_{\sigma}$  and  $F_{\pi}$ , according to the theory of Darwin and for the case of negligible absorption, are given by,

$$F_{\sigma}(\ell) = \begin{cases} \left[ \frac{F/A}{\ell - \sqrt{\ell^2 - F^2/Z^2}} \right]^2 & \text{when } \ell < -F/Z \\ 1 & \text{when } -F/Z < \ell < F/Z \\ \left[ \frac{F/Z}{\ell + \sqrt{\ell^2 - F^2/Z^2}} \right]^2 & \text{when } \ell > F/Z \end{cases}$$



$$= \left[ \frac{F/Z}{\ell - \sqrt{\ell^2 - F^2/Z^2} \cos^2 2\theta_0} \right]^2 \quad \text{when } \ell < -(F/Z) \cos 2\theta_0$$

$$F_{\pi}(\ell) = 1 \quad \text{when } -(F/Z) \cos 2\theta_0 < \ell < (F/Z) \cos 2\theta_0$$

$$= \left[ \frac{F/Z}{\ell + \sqrt{\ell^2 - (F^2/Z^2) \cos^2 2\theta_0}} \right]^2 \quad \text{when } \ell > (F/Z) \cos 2\theta_0$$

$$\text{where } \ell = (\theta - \theta_B) \sin 2\theta_0 / 2\delta - 1$$

F is the crystal structure factor

Z is the total number of electrons per unit cell

$\delta = 1$  - index of refraction

$\sigma$  and  $\pi$  are defined in footnote page 18.

It has been shown in Compton and Allison<sup>(8)</sup> p. 397 that

$$\int_{-\infty}^{\infty} F_{\sigma}(\ell) d\ell = \frac{8F\delta}{3Z \sin 2\theta_0}$$

$$\text{and } \int_{-\infty}^{\infty} F_{\pi}(\ell) d\ell = \frac{8F\delta \cos 2\theta_0}{3Z \sin 2\theta_0} \quad (63)$$

However, in eq. (62) the arguments of  $F_{\sigma}$  and  $F_{\pi}$  are not independent but are related through eqs. (41) and (40), so that the above integrals do not apply. Attempts to integrate (62) analytically have failed.

To have some idea as to the intensity reflected by the two crystals, let us assume that  $C_{\pi} = 1$ ,  $\lambda = \lambda_0$ ,  $z = \eta = 0$ , that is, we shall consider surface reflections from the centers of the two crystals for one wavelength only. Eqs. (41) and (40) then become,

$$\theta - \theta_B = t$$

$$\theta' - \theta_B = t'$$

where  $t$  and  $t'$  are independent variables ranging from  $+1/1500$  rad. to  $-1/1500$  rad. or  $144''$  to  $-144''$ . Since the functions  $F_{\sigma}$  and  $F_{\pi}$  fall quite rapidly outside the range of complete reflection, it is justifiable to set the ranges of integration on  $t$  and  $t'$  from  $\infty$  to  $-\infty$ .

Setting  $J(\lambda - \lambda_0) = 1$ , we have, therefore,

$$I = (1/2)I_0 \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} [F_{\sigma}(t)F_{\pi}(t') + F_{\pi}(t)F_{\sigma}(t')] dt dt'$$

and in view of eqs. (63)

$$I = I_0 \left[ \frac{8F_{\sigma}}{3Z \sin 2\theta_0} \right]^2 \cos 2\theta_0 \quad (64)$$

The loss in intensity due to polarization manifests itself in the term  $\cos 2\theta_0$  which is due to the function  $F_{\pi}$ . Eq. (64), though not applicable to the crystals as a whole, nevertheless gives a fair indication as to the reflection ability in the neighborhood of their centers. This equation can be used as a criterion for comparison with other crystals, and suggests the use of a crystal for which the values of  $F/Z$ ,  $\mathcal{J}$  and  $\cos 2\theta_0$  are larger than the corresponding values in quartz. Although there are numerous crystals that satisfy the above conditions, there are very few that are sufficiently large in size and have the necessary elastic properties for bending. A search of the literature has revealed only one crystal that is comparable to quartz in strength, namely topaz. It will be of interest to compare the properties of quartz and topaz.

	<u>Quartz</u>	<u>Topaz</u>	
Formula	SiO <sub>2</sub>	[Al(F,OH)] <sub>2</sub> SiO <sub>4</sub>	
Planes used	(310)	(303)	(006)
Grating spacing, d	1.178 <sup>0</sup> Å	1.352 <sup>0</sup> Å	1.395
Molecules/unit cell	3	4	4
Electrons/unit cell, Z	90	360	360
Volume of unit cell, V	112 <sup>0</sup> Å <sup>3</sup>	341.5 <sup>0</sup> Å <sup>3</sup>	341.5 <sup>0</sup> Å <sup>3</sup>
Crystal structure factor, F	21.8	139.0	124.4

From Compton and Allison<sup>(8)</sup> p. 280

$$\sigma = \frac{(Z/V)e^2 \lambda_o^2}{2\pi mc^2} \quad (65)$$

where  $e^2/mc^2$  is the classical radius of the electron ( $2.817 \times 10^{-5}$ Å), and  $\lambda_o = 1.537$ Å.

The Bragg angle is given by

$$\theta_o = \sin^{-1}(\lambda_o/2d) \quad (66)$$

We can now substitute numbers into (66), (65) and (64) and get

	<u>Quartz</u>	<u>Topaz</u>	
Planes	(310)	(303)	(006)
$\theta_o =$	40.7°	34.6°	33.4°
$\cos 2\theta_o =$	0.1497	0.3555	0.3940
$\sigma =$	$0.85 \times 10^{-5}$	$1.12 \times 10^{-5}$	$1.12 \times 10^{-5}$
$F/Z =$	0.242	0.386	0.346
$I/I_o$	$4.6 \times 10^{-12}$	$54 \times 10^{-12}$	$50 \times 10^{-12}$

One can, therefore, expect an increase in beam intensity (at the surface) by replacing the quartz crystals with topaz

of about  $54/4.6 = 12$  times or  $50/4.6 = 11$  times depending on the planes used.

Since it is possible to obtain reflections from planes within the crystals as well, it is necessary to take into account the function  $C_{\frac{\sigma}{\pi}}$ . We shall still restrict ourselves to the centers of the crystals. The function  $C$  is given by (11)

$$C_{\frac{\sigma}{\pi}} = \left\{ 1 - \exp \left[ - \mu T (\sin(\theta_0 + \alpha) + \sin(\theta_0 - \alpha)) / \sin(\theta_0 + \alpha) \sin(\theta_0 - \alpha) \right] \right\} \cdot \left\{ 1 - \exp \left[ - \epsilon_{\text{ave}} w_{\frac{\sigma}{\pi}} R / \cos \theta_0 \cos \alpha (1 + \sin^2 \alpha) \right] \right\} \cdot \left\{ 1 - \exp \left[ - \mu w_{\frac{\sigma}{\pi}} R (\sin(\theta_0 + \alpha) + \sin(\theta_0 - \alpha)) / \sin(\theta_0 - \alpha) \cos \theta_0 \cos \alpha (1 + \sin^2 \alpha) \right] \right\} \quad (67)$$

where  $\mu$  = linear absorption coefficient

$T$  = thickness of the crystals = 0.07 cm.

$R$  = radius of curvature = 120 cm.

$\epsilon_{\text{ave}}$  = average extinction coefficient

$\alpha$  = angle between crystal planes and faces

$w_{\frac{\sigma}{\pi}}$  = diffraction pattern widths

Although eq. (67) is supposed to express the area under a rocking curve it is applicable here. In the case of a rocking curve experiment, a fine parallel beam of x-rays strikes a crystal which is rotated a few seconds of arc at a time, the reflected beam being measured in each position. In our case, because of the finite size of the target, rays from different parts of it converge and strike the center of the crystal with different grazing angles so that the total intensity is equal to the area under the rocking curve mentioned in the previous case.

We again compare quartz and topaz. However, it is first necessary to compute  $\mu$ ,  $\epsilon_{ave}$  and  $w_{\sigma/\pi}$ .

$$\mu = g \sum_i p_i \mu_i \quad \text{where } g = \text{density of crystal}$$

$p_i$  = mass of element  $i$  in the crystal/total mass

$\mu_i$  = mass absorption coefficient of element  $i$

$$\epsilon_{ave} = \frac{3\pi e^2 \lambda_o F}{8 mc^2 V} = 5.09 \times 10^3 (F/V) \text{ cm}^{-1}$$

$$w_{\sigma} = (4/3) 4\delta (F/Z) \csc 2\theta_o$$

$$w_{\pi} = (4/3) 4\delta (F/Z) \cot 2\theta_o$$

where the factor  $4/3$  is applied to the actual width in order to approximate the actual diffraction pattern by a rectangular pattern of the same area. This approximation has been used in deriving eq. (67).

The composition and specific gravity of the two kinds of crystals are given in the following table:

	<u>Quartz</u>	<u>Topaz</u>	<u>Mass absorption coefficient <math>\mu_i</math></u>
Composition, $p_i$	Si 41.7% O 53.3%	Si 15.17% O 34.67% Al 29.58% F 20.58%	Si 60.0 O 11.16 Al 49.0 F 17.0
Specific gravity	$g = 2.65$	3.53	

from which the values of  $\mu$  can be calculated.

	<u>Quartz</u>	<u>Topaz</u>	
Planes	(310)	(303)	(006)
Linear absorption coefficient	$\mu = 82 \text{ cm}^{-1}$	$109 \text{ cm}^{-1}$	$109 \text{ cm}^{-1}$
Average extinction coefficient	$\epsilon_{ave} = 990 \text{ cm}^{-1}$	$2070 \text{ cm}^{-1}$	$1850 \text{ cm}^{-1}$
Diffraction pattern widths	$w_{\sigma} = 1.112 \times 10^{-5}$ $w_{\pi} = 0.166 \times 10^{-5}$	$2.462 \times 10^{-5}$ $0.875 \times 10^{-5}$	$2.254 \times 10^{-5}$ $0.886 \times 10^{-5}$

Substituting the above values into eq. (67) together with  $T = 0.07$  cm. and  $\alpha = 2.1^\circ$ , it is seen that the argument of the first exponential term is very large, so that the first bracketed term is equal to unity. This indicates that our crystals are sufficiently thick and that the function  $C$  does not depend on their thickness.

Hence, for the two kinds of crystals and two types of polarization, we have

		<u>Quartz</u>	<u>Topaz</u>	<u>Ratio, Topaz to Quartz</u>		
			(303)	(006)	(303)	(006)
$\sigma$ polarization	$C_\sigma = 3.12$	1.78	1.90	0.57	0.61	
$\pi$ polarization	$C_\pi = 5.19$	3.65	3.61	0.70	0.70	

The above calculations are based on reflections from a single bent crystal. When applied to the two crystal monochromator, the above ratios must be multiplied by one another since the  $\sigma$  polarization for one crystal is the  $\pi$  polarization for the other and vice versa. Although these ratios are unfavorable, the factors of 12 and 11 obtained previously, more than compensate for them. When the latter are taken into account, the gain in intensity with topaz is

4.8 for the (303) planes.

4.7 for the (006) planes.

It must be remembered that the above calculations were carried out for the centers of the crystals only and may not be valid for the whole crystals. However, on the basis of experimental work conducted by Ingelstam<sup>(12)</sup>, the actual gain in intensity

from bent topaz crystals is not far different from the above figures.

Other means of increasing the intensity consist in enclosing the beam in helium, and using a beam which emerges with a smaller grazing angle from the target. Since the total path length of the beam is about 150 cm., the loss due to absorption in air is considerable. The substitution of helium for air improves the intensity by a factor of 3.5. The beam intensity at a smaller angle of emergence is about 2.5 times as great per unit target area. However, the projected target area is reduced by about a factor of five. Since the useful target area is a strip no larger than 0.2 mm. wide, the use of such a beam is justifiable. Taking the above factors (substitution of topaz for quartz, use of helium atmosphere, and reorientation of x-ray tube) into consideration, one can, therefore, expect a gain of 42 in intensity in the case of the (303) planes of topaz or of 41 in the case of (006) planes over the intensity available before making these changes.

PART IV

DESCRIPTION OF THE POINT FOCUSING

MONOCHROMATOR

The problem of orienting two bent crystals correctly in space involves twelve independent degrees of freedom. It is not hard to see then, that unless some method is devised for lining up the crystals, in a logical or systematic manner, the solution to the problem by cut-and-try adjustment is next to impossible. The instrument which we shall describe below was designed and built with this problem in mind.

The instrument is built mainly out of steel. To avoid as much as possible any unpredictable lining up problems, all defining surfaces are surface ground with the exception of the pads on the triangular table. The cylindrical faces of the crystal clamping blocks (approximately 120 cm. in radius) are both ground and lapped by a special method described by DuMond, Lind and Cohen<sup>(13)</sup>. This shop method has the advantages that it automatically insures: (1) that the generators of the precision cylindrical clamping surfaces shall be truly normal to plane ground reference faces on the top and bottom of the crystal clamping blocks and, (2) that the radii of curvature on blocks for crystals A and B shall be closely equal. All welded parts were heat treated to prevent distortion in time.



Because of its three dimensional character, the instrument and its lining up procedure are best described by a series of plates.

PLATE 1

This is an overall view of the instrument with the parts assembled for use except that the crystals and the front halves of the crystal clamping blocks have been removed. It consists of a triangular steel table mounted by means of leveling screws on three concrete pillars. The table is provided with a number of pads which are welded to it and which are machined accurately so as to define a common reference surface. The crystal laminae are cylindrically bent by clamping between pairs of steel crystal bending blocks of which only the rear members are shown in the photograph. The block with vertical generators (crystal A) is mounted on a horizontal radius arm (R), while the block with horizontal generators (crystal B) is mounted on a radius arm (R') in the vertical plane. The area of the rectangular hole in the block marked (A) is approximately the effective crystal area. (See Plate 6 for a close-up view of the crystal blocks with crystals clamped between front and back members.) The vertical structure in Plate 1 supports the detecting device at the top (P) and the scattering sample at the bottom (S). The detecting device at present is a film holder, but provision is made for a Geiger counter which will be used when a beam of sufficient intensity is attained.

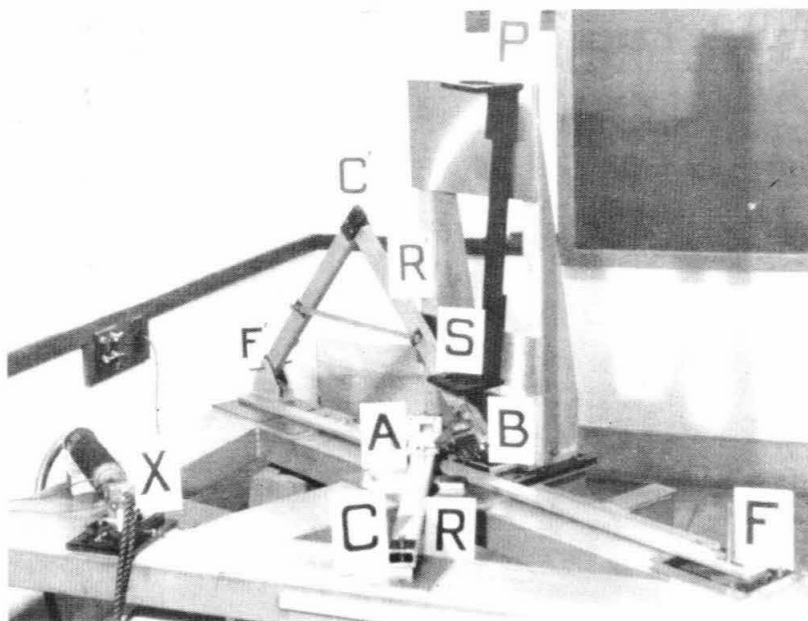


PLATE 1 - This is an overall view of the instrument with the x-ray tube X on the left, crystal blocks (rear halves) A and B at the center, and the point focus P at the top.

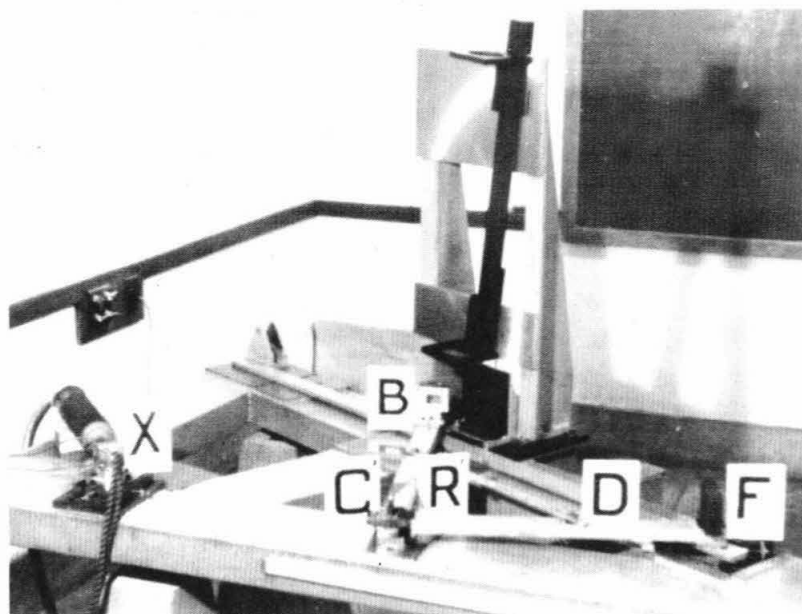


PLATE 2 - For the purpose of alignment, crystal B, with its associated radius arms R' and D, is mounted in the horizontal position as shown, R' and D, being free to turn independently about C'.

The beam diverges from the x-ray tube (X) and proceeds to crystal A, where it is Bragg-reflected ( $\text{CuK}\alpha_1$ ), so that in the absence of crystal B, it would come to a vertical line focus at F. However, crystal B intercepts the beam and by Bragg reflection, in the vertical plane, brings the twice reflected beam to a point focus at P. We shall refer to the centers of the reflecting surfaces of crystals A and B simply as the points A and B.

The points X, A, and F lie on the horizontal focal circle (of crystal A), whose center is at C. The points F', B and P lie on the vertical focal circle (of crystal B), whose center is at C'. The two focal circles are congruent, with point F' corresponding to F, B to A, and P to X.

The long bar FF' pivots about a vertical axis at F, carrying with it the vertical structure and the assembly supporting crystal B. Hence the horizontal and vertical focal circles can be adjusted in such a way as to maintain the points F', A, B, and F in a straight line which is also the line of intersection of the planes of the two focal circles.

The vertical pin at F supports a film holder which allows first crystal B and then crystal A to be individually aligned photographically, while in the horizontal position. This alignment process is made clearer in the following plate.

#### PLATE 2

For the purpose of alignment, crystal B, with its associated radius arms R' and D, is mounted in the horizontal position

as shown, R' and D being free to turn independently about C'. The top of the crystal block is made parallel to the reference surface by means of a 1/10,000 inch dial indicator. This adjustment takes care of two rotational degrees of freedom of crystal B.

Next, a narrow line source of light, with a microscope focused to a point right above it is placed in line with C' and B. Crystal B is pivoted about a vertical axis, passing through B, until a line image due to the reflected light from the crystal face, appears right above the light source and hence in the field of view of the microscope. The light source and microscope are then moved along the line C'B until the image appears to be the sharpest. By measuring the distance between the light source and the center of crystal B, the diameter of the focal circle is determined. This permits us to set crystal B at the right distance from C', namely, half the focal diameter. This concludes the two optical adjustments which take care of one more rotational degree of freedom, making a total of three. The preceding statement may seem to be slightly contradictory, but it will be shown to be true by the concluding statement of the next paragraph.

The arm R' is then pivoted about C' until the line focus, due to Bragg reflection of the  $\text{CuK}\alpha_1$  line, appears on the film, in the cassette at F. The axis of the pin at F is then moved laterally to coincide with the line focus. Since one

end of the radius arm D is constrained to move with F, the angle between D and R' has thus been established, and the two arms are now rigidly clamped together by means of a cross-bar. This assembly is then removed and placed aside to permit alignment of crystal A, shown on the next plate. After crystal A is permanently adjusted in place, there will remain only the three translatory degrees of freedom of crystal B relative to A, which for convenience will be broken down into two rotations and one translation.

PLATE 3

In this photograph, crystal A and its radius arm are shown in their normal positions. The dial indicator and optical instruments are again used to orient the crystal correctly on its radius arm. If crystal A is identical to B, then the point F should remain fixed in position and the only adjustment is the location of point A relative to F\*. This is done photographically by moving radius arm R about C, until the line focus appears at F. The arm is then clamped permanently in place and the assembly supporting crystal B is then mounted as shown in the next plate. As was mentioned before, the correct placement of crystal A eliminates six more degrees of freedom, thus making a total of nine eliminated so far.

---

\* It was found, however, that the radii R and R' differed by about 6 mm, hence requiring a corresponding change in the length CF. It is hard to say how this slight difference in radii actually affects the point focus.

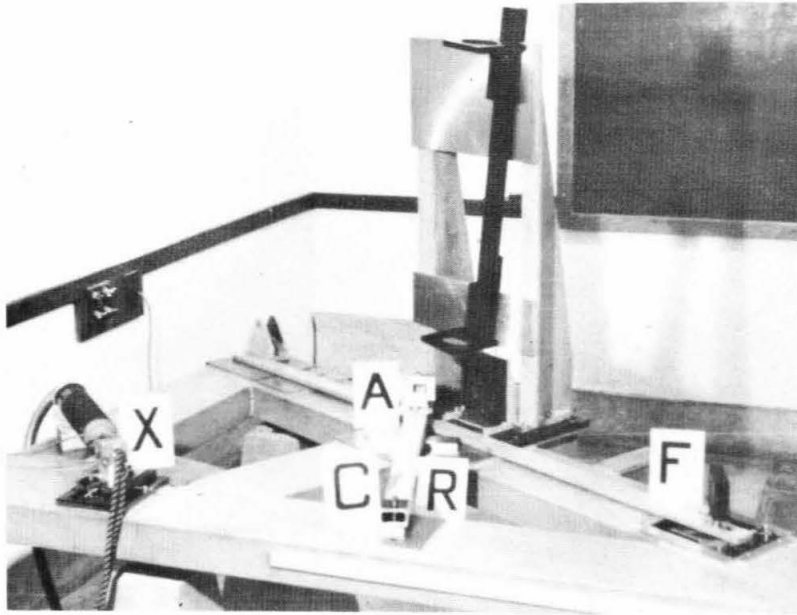


PLATE 3 - Crystal A and its radius arm R are lined up in their normal positions as shown.

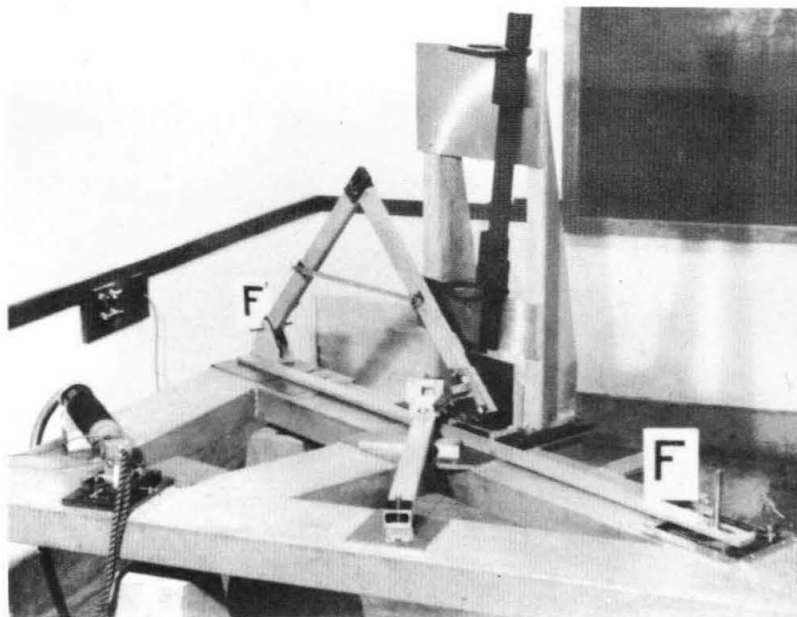


PLATE 4 - The last adjustments consist of lining up points F and F' with the center of crystal A, correcting for astigmatism and ascertaining that crystal B is set at the Bragg angle to accept radiation from A.

PLATE 4

The only adjustments left now are: (1) to line up the points F and F' with A, (2) to correct for astigmatism and, (3) to ascertain that crystal B is set at the Bragg angle to accept radiation from A. The latter adjustment is made by pivoting crystal B with its assembly about the horizontal rod at F'. The first adjustment is made by connecting two points directly above F and F' with a tight-line and pivoting the long bar FF' about the vertical rod at F until the tight-line is right above the point A. The actual point A is inaccessible, so that a scratch mark on the crystal block directly above it is used instead. Astigmatism is corrected by moving the bar FF' longitudinally, thus pulling crystal B toward or away from crystal A until the point P appears sharply defined. This adjustment is not particularly sensitive. Moving the rod one centimeter one way or another doesn't seem to make much difference.

The three adjustments, just described, eliminate the last three degrees of freedom thus concluding our lining up procedure.

PLATE 5

This plate shows the complete instrument with the exception of the lead partitions, one in front of crystal A and one between the two crystals as well as the lead box which surrounds both crystals. The partitions, consisting of a lead sheet with  $3/4$ " x  $3/4$ " square windows to let the beam through,

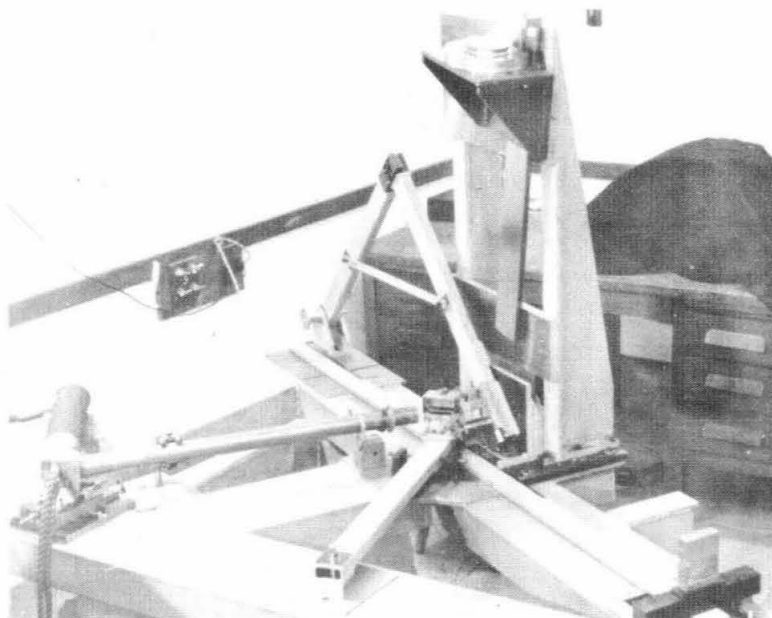


PLATE 5 - The complete instrument with the exception of the lead housing surrounding the two crystal blocks.

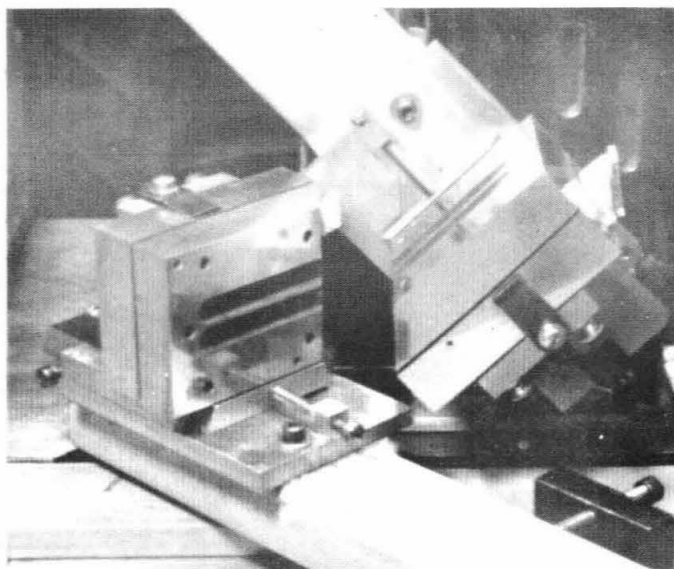


PLATE 6 - Close-up view of the crystal blocks shown with the various adjusting screws.



are used to cut down the incoherent scattering as well as fluorescent radiation from the steel crystal blocks, thus reducing considerably the background in the neighborhood of the point focus. To prevent scattering from the steel webs of the crystal blocks, the latter are covered with thin lead foil. The lead box protects the occupants of the room from irradiation. The x-ray tube is a diffraction type tube made by the Machlett Company and is driven by a North American Philips basic diffraction unit which can deliver continuously 60 PKV at a full wave rectified current rating of 50 ma. The optimum operation of the tube, however, is at 35 PKV and 20 ma. The tube that extends from the x-ray tube to crystal A is filled with helium, thus cutting down loss of intensity due to air scattering. At the top of the vertical structure one can observe the film holder mounted on an angle plate which can ride up and down on ways along the beam thus permitting adjustment of the x-ray film to coincide with the best point focus. The scattering sample is mounted on a holder, not here shown, which can also ride up and down along the beam. Every ray which passes through the sample will be partly scattered into a narrow cone of diffusion around said ray. The resulting diffraction pattern in the focal plane is the superposition of all such scattered rays; the undeviated rays which come to a point focus being suppressed by means of a thin tungsten wire (0.020" in diam.) placed against the photographic plate. With this arrangement, every point on a circle of specified radius

concentric with the central focal spot in the focal plane receives radiation which has been scattered by every portion of the scatterer under approximately the same angle. Thus, aside from a small cosine correction coming from the fact that the rays are not parallel, the diffraction pattern represents truly the scattering property of the sample.\*

#### THE CRYSTALS AND CRYSTAL BLOCKS

##### PLATE 6

This is a close-up view of the crystal blocks A and B. One can observe some of the local adjustments necessary to set crystals A and B correctly on their respective radius arms R and R'.

The blocks are made of a special type of stainless steel which is quite stable and does not distort with time. This is quite important since the curved surfaces must retain their optical properties with high precision. In order to insure uniformity of curvature, the two convex blocks were mounted on a steel plate and treated as a single unit, that is, they were cylindrically ground and lapped simultaneously. The same was done with the concave blocks. The method of grinding referred to on page 53 is shown in plates 7 and 8.

##### PLATES 7 and 8

The convex face (on the front half of the crystal block)

---

\*This statement is ideally true. However, since the point focus has a finite size, a small correction must be made in regions very close to the center of the diffraction pattern.

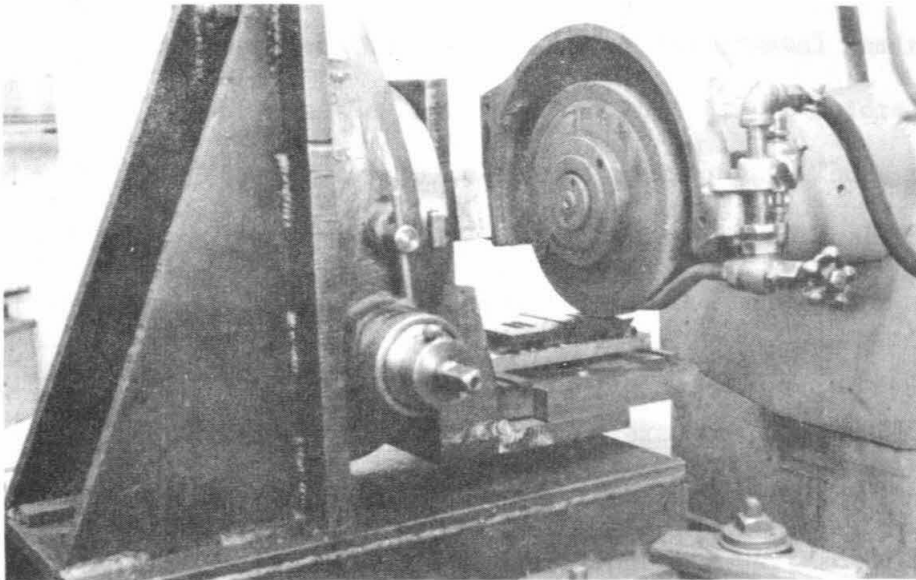


PLATE 7 - Method of generating the precision cylindrical surfaces of the curved crystal clamping blocks for the point-focusing x-ray monochromator in an ordinary flat surface grinder. The two stainless steel blocks are seen mounted on the rocking bracket under the grinder wheel. The bracket, mounted on a pivot, rocks to and fro as the work travels backward and forward on the transverse ways of the grinder. The rocking is caused by a lever arm rigidly clamped to the bracket whose far end is provided with a roller which rolls along an inclined straight-edge in unison with the transverse carriage movement

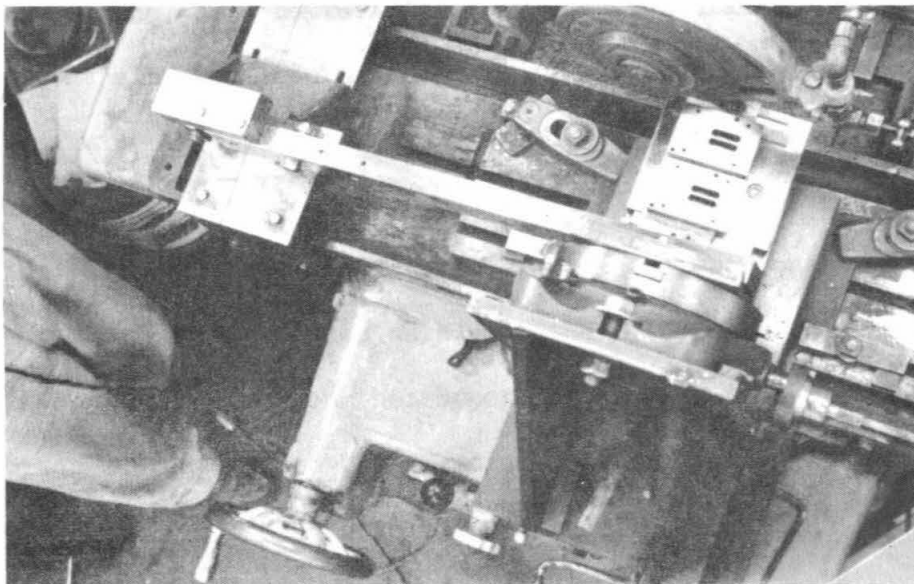


PLATE 8 - View (from above) of the method of precision profiling the cylindrical surfaces of the above mentioned clamping blocks in a flat surface grinder. The blocks can be seen mounted on the rocking bracket. The long lever arm with the small roller at its left-hand end is here clearly visible. The weight attached to the left end of the lever arm to hold the roller in contact with the plane on which it rolls can also be seen.

forms the defining surface against which the crystal lamina is bent. The crystal is backed by a rubber gasket before the rear half of the crystal block is clamped on. Thus, only the convex face must be accurately lapped to an optical finish. At the end of the lapping procedure described in reference 13, the concave cast iron lap, which itself acquires a high optical polish, is set on an optical bench and examined optically by placing a line source of light at the center of curvature and observing its image with a microscope. A Hartman diagram of two zones reveals that the focus is approximately 0.016 mm. wide. This, then, indicates the degree of optical perfection attained by the convex crystal blocks.

The crystal laminae were cut from a single slab of quartz, which was ground flat and polished to approximately the final thickness prior to cutting.

The angle between the (310) crystal planes and the faces of the slab was determined on the two crystal spectrometer by replacing crystal B thereon with the slab (set for reflection of the beam from the above mentioned (310) planes) and locating the "parallel position". Crystal A was another quartz plate whose reflecting planes were also the (310) planes. The slab (whose orientation on the support bracket of the crystal B pivot was determined by a 3-point contact between the polished crystal surface and three ball bearings pressed tightly into shallow holes in the surface of the bracket) was then rotated  $180^\circ$  about an axis perpendicular to its face and the "parallel

position" was again located. The difference in the two "parallel position" readings is equal to twice the angle between the crystal planes and its faces. This angle was re-adjusted, by grinding, to the desired value  $\alpha$ , and by keeping track of its direction, it was possible to clamp the crystals, when finished, correctly in the crystal blocks. (See page 9 for the requirement which fixes  $\alpha$ .)

Special precautions were taken in lapping to prevent the development of surface stresses. Unnecessary heating in the lapping procedure was avoided. In the final stages of lapping, only the finest abrasive was used. The prolonged use of the fine abrasive made certain that no scratches were left from the coarser abrasive. Such scratches are invisible but can be revealed by etching the quartz. This is based on the theory that the scratches are actually grooves filled up with minute particles of quartz which can be removed by etching. The minute particles wedged in the grooves contribute to the surface stresses. Thus, if the two faces of a lamina are not scratched equally (which is usually the case), the etching process which relieves the strain will tend to make the lamina "curl" thus ruining the optical features of its faces. It is necessary to etch the crystals for two reasons: (1) to prevent breaking when being bent due to surface stresses, (2) to relieve the surface strains in order to be sure that the crystal planes are actually plane prior to bending.

The crystals used in the point-focusing monochromator were etched and found to be without a single scratch. Furthermore, they were examined on an optical flat before and after etching and no appreciable change was observed. The crystals are optically flat to within two fringes, the slight curvature tending to change the radius of the focal circle slightly.

## PART V

### EXPERIMENTAL RESULTS

Having obtained a focused beam by the procedures of Part IV, the problem is to find some appropriate sample for a scattering experiment. Since none of the improvements of beam intensity suggested in Part III were made at the date of writing this thesis, it was necessary to limit oneself to a relatively good scatterer. Two different samples of carbon black were picked out, one with average particle radius  $54 \text{ \AA}$  (carbolac 1) and the other with radius  $230 \text{ \AA}$ . The size and size distribution of the former was given by the manufacturer on the basis of electron microscope measurements and the latter was determined from the measurement of ninety particles photographed on the electron microscope in the Chemistry Division.

Carbon blacks, including the above, have been studied before<sup>(14,15,16)</sup>, so that the present study does not reveal anything new regarding these samples. However, the purpose of the experiment was to check on the performance of the instrument and, particularly, on the luminosity of the direct beam. Since there exists no simple way to measure the direct beam intensity, the exposures required for satisfactory diffraction patterns from the carbon blacks are a good measure of the performance of the point focusing instrument.

The sample of carbon black was held in a ring 5 cm. in diameter and 0.4 cm. in axial thickness, with a 0.001" thick

nylon sheet glued to each side (to form a shallow cylindrical cavity which was packed full of the carbon). It was placed at a distance of 655 mm. from the point focus where the photographic plate was located. The irradiated volume was approximately  $1.6 \text{ cm}^3$ .

#### METHOD OF DETECTION

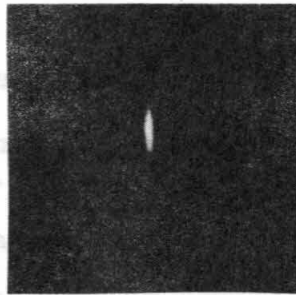
As has been previously mentioned, the scattered intensity was recorded on x-ray film, the direct beam being blocked out by means of a tungsten wire. See Plate 9.

After the diffraction pattern was obtained, it was necessary to measure the film density (and hence, the scattered intensity) versus the distance away from the center i.e. the scattering angle. This was done by means of a recording microphotometer. The light beam of the microphotometer was approximately the same elongated shape and size (0.89 mm. x 0.18 mm.) as the point focus and was oriented in the same way relative to the x-ray film. This light beam was allowed to scan the diffraction pattern along a diameter, the transmitted light being allowed to illuminate one junction of a thermopile which was connected to the coil of a d'Arsonval galvanometer, and the deflection of the latter being recorded photographically.

We have assumed that the following formula holds true, namely,

$$I_s = (1/kt) \log (x_0/x) \quad (68)$$





- (a) Point Focus (Enlarged 5.8 times)  
3 minute exposure at 20 PKV and 20 MA with beam in air. The half-width of the point represents a scattering angle of about  $40''$ , with scatterer 65 cm. away.

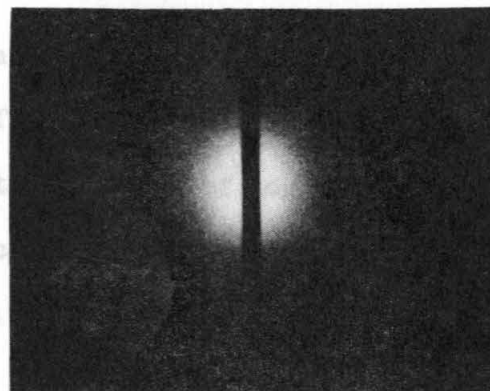
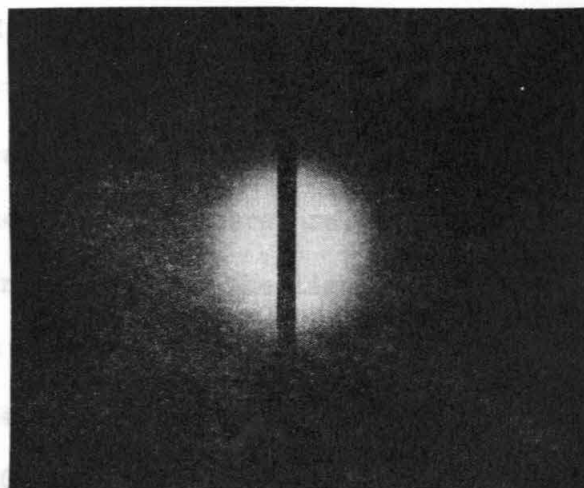


PLATE 9

- (b) Diffraction Pattern of Carbon Black (Carbolac 1)  
(Enlarged 5.8 times)  
3 hours exposure at 35 PKV and 20 MA with beam in helium atmosphere. Average particle radius 54 Å.



- (c) Diffraction of Carbon Black (Enlarged 5.8 times)  
3 hours exposure at 35 PKV and 20 MA with beam in helium atmosphere. Average particle radius 230 Å.

where  $I_s$  is the scattered x-ray intensity.

$k$  is some constant dependent on the speed of the x-ray film.

$t$  is the x-ray exposure time.

$x_0$  is the maximum galvanometer deflection from totally dark film to clear film,

$x$  is the galvanometer deflection from totally dark film to some exposed portion of the film.

This formula and its range of validity were verified experimentally.

In the case of small particles, theoretical curves for various types of particle size distributions have been worked out<sup>(4)</sup> and plotted in terms of  $\log (I_s/I_0)$ . It is easily seen that the coefficient of the logarithmic term above appears as an additive constant in such a plot and is, therefore, of little interest.

The microphotometer curve is treated in the following manner. It is placed over a sheet of ordinary graph paper and the values  $x$  are read off for various scattering angles on both sides of the maximum. The dark readings having been taken at the beginning and the end of the microphotometer run are connected by a straight line and used as a reference line from which  $x$  is measured. Although the thermopile drifts during the run due to increase in temperature, the value of  $x_0$  remains almost constant since both the light and dark readings are affected in the same way.

A commercially built microphotometer with beam size 0.01 mm. x 0.05 mm. was also used to analyze the diffraction

patterns but no appreciable difference was observed in the microphotometer runs of the two instruments. Microphotometer curves by the latter instrument for the two kinds of carbon blacks are shown in Figs. 9 and 10.

The ratio  $x_0/x$  is calculated and its logarithm is plotted versus the square of the scattering angle on log paper. Curves representing different exposures for a given sample are plotted on the same graph and are then compared with theoretical curves worked out by Roess and Shull<sup>(4)</sup>. The method of comparison is explained in the section that follows.

#### LOW ANGLE SCATTERING DUE TO SMALL PARTICLES

The above mentioned investigators have worked out the intensity curves for spherical particles with different types of size distributions, rectangular, Maxwellian and Gaussian. The Maxwellian type distributions seem to be the only applicable ones to our carbon blacks, see Fig. 11.

The theory upon which the above curves are based can be summarized in the following manner. The intensity scattered at a small angle  $\epsilon$  by particles whose size distribution is  $M(R)$ ,  $R$  being the radius, is given by

$$I(\xi) = KI_e \rho_e^2 \int_0^\infty M(R) R^3 S(R, \xi) dR \quad (69)$$

where

$$\xi = (2\pi/\lambda_0)\epsilon \quad (70)$$

$I_e$  is the Thomson scattering factor which is essentially constant for the small angles we shall consider.  $\rho_e$  is the electron density assumed uniform throughout the particle and  $S(R, \xi)$  is the

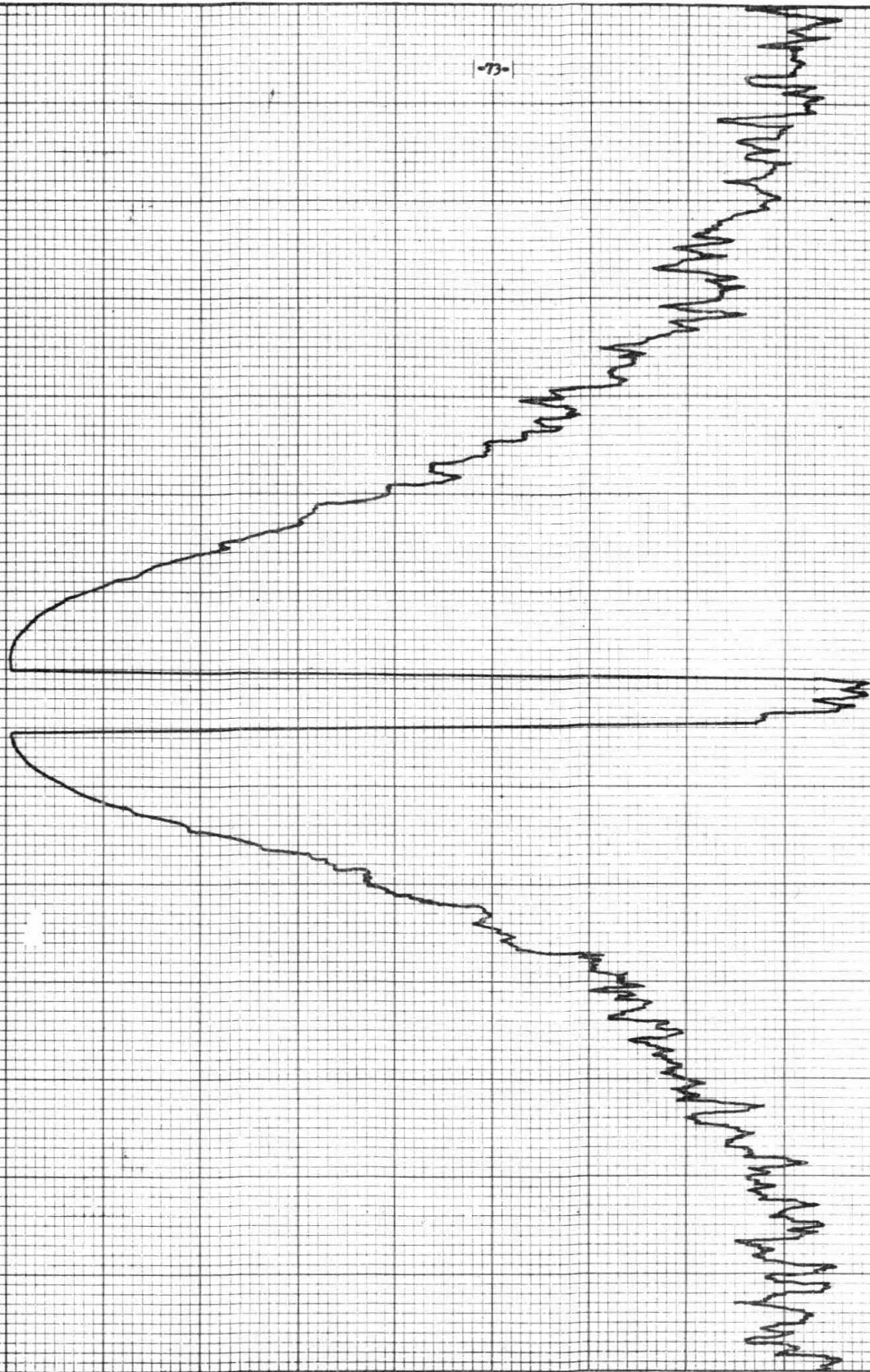


Fig. 9 - Microphotometer curve of diffraction pattern shown in Plate 9 (b).  
Intervals at the bottom represent 0.50 mm. on diffraction pattern.

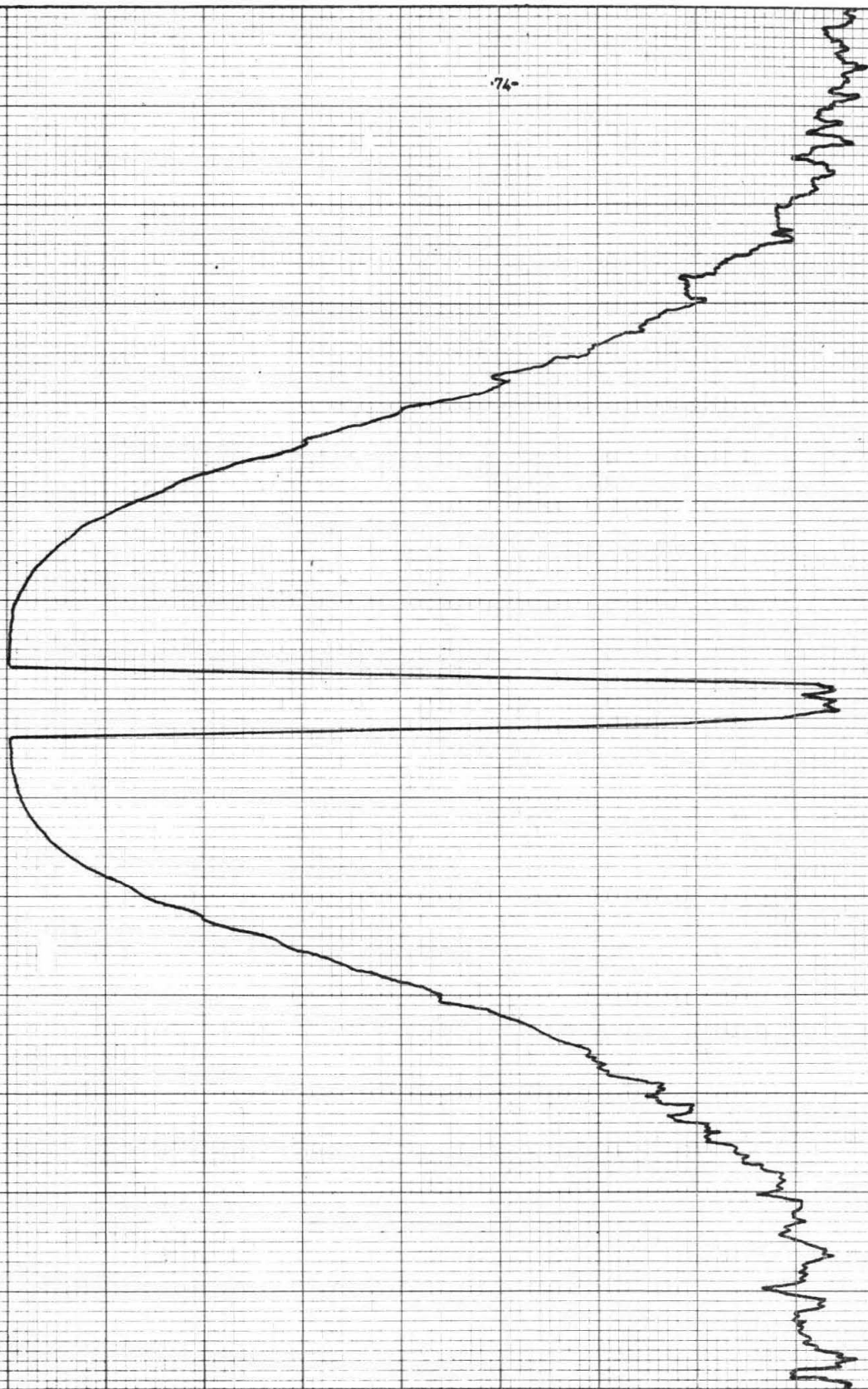


Fig. 10 - Microphotometer curve of diffraction pattern shown in Plate 9 (c).  
Intervals at the bottom represent 0.50 mm. on diffraction pattern.



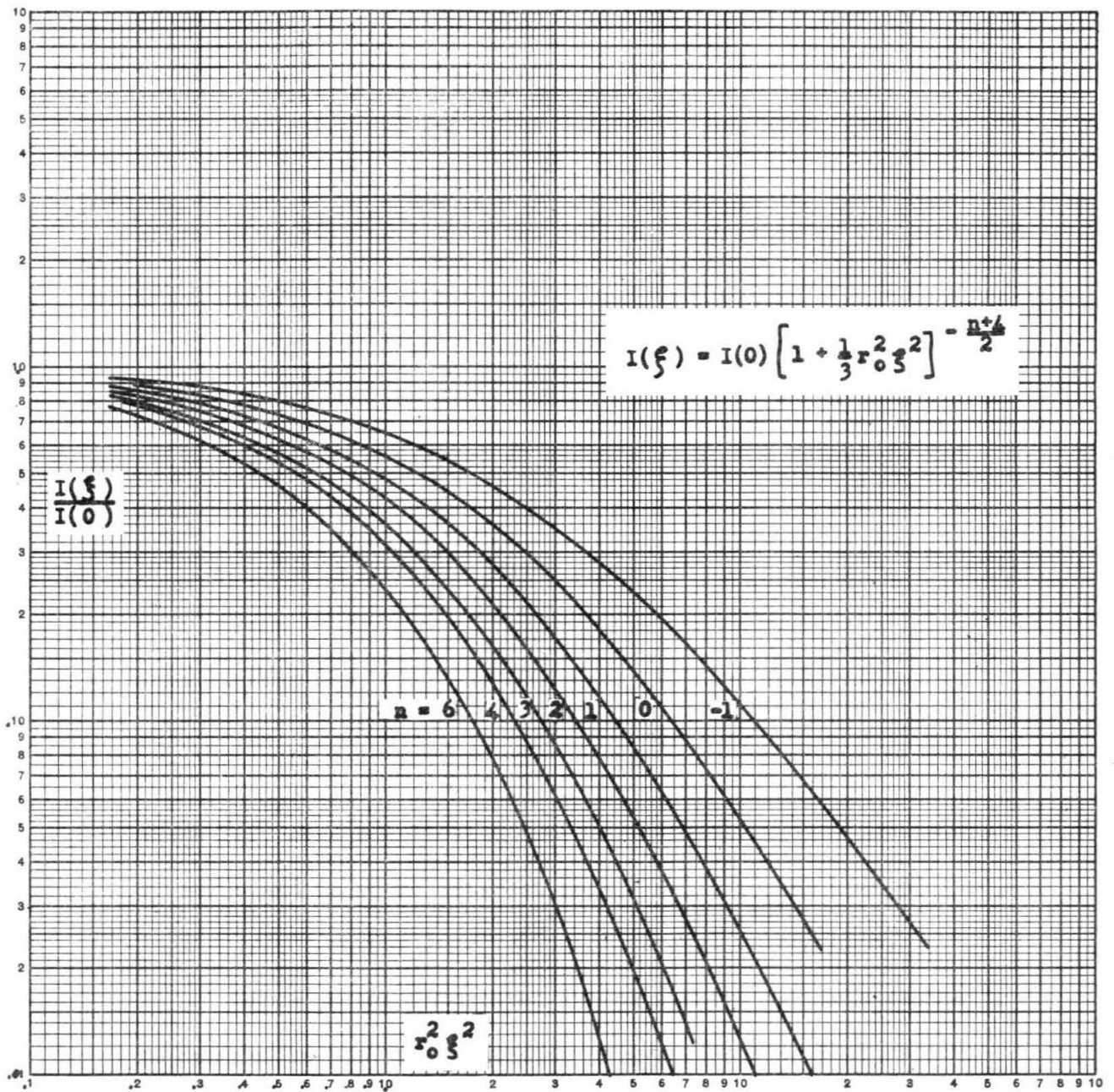


Fig. 11 - Angular dependence of scattered intensity for Maxwellian distribution of particle sizes.

scattering function characteristic of the particle size  $R$  and its geometrical shape.  $M(R)dR$  represents the total mass of particles in the size range  $R$  to  $R+dR$ .  $K$  is a constant proportional to the total mass.

For spherical particles Guinier<sup>(1)</sup> has shown that

$$S(R, \xi) = \exp(-r^2 \xi^2/3) \quad (71)$$

where he identifies  $r$  with the radius of gyration of the particle.

For spheres

$$R = (5/3)^{1/2} r \quad (72)$$

The Maxwellian distribution function  $M$ , which we are interested in, can be expressed as a function of the radius of gyration and the parameters  $r_0$  and  $n$  as follows

$$M(r) = \frac{2(r/r_0)^n}{r_0 \Gamma(\frac{n+1}{2})} \exp(-r^2/r_0^2) \quad (73)$$

Substituting eqs. (71) and (73) into (69) and integrating, we

get

$$I(\xi) = I(0) \left[ r_0^2 \xi^2 + 1 \right]^{-(n+4)/2} \quad (74)$$

where  $I(0)$  is a complicated expression involving the parameters  $r_0$  and  $n$  and is of no interest here. Eq. (74) has been plotted,  $\log I(\xi)/I(0)$  versus  $\log r_0^2 \xi^2$ , for several values of  $n$ .

Since we are interested in the average particle radius  $\bar{R}$ , we can define the average radius of gyration  $\bar{r}$  as that radius which divides the mass distribution  $M(r)$  in two. This has been worked out<sup>(4)</sup> for several values of  $n$ , as follows:

$n$	0	1	2	3	4	5
$\bar{r}/r_0$	0.227	0.693	1.183	1.677	2.176	2.674

The experimental points are plotted  $\log\log(x_0/x)$  versus  $\log \xi^2$ . By translating the experimental points horizontally and vertically until they match one of the theoretical curves, the value  $n$  is obtained immediately. If the horizontal shift of the experimental points relative to the theoretical curve is  $x$  units, this being obtained by dividing any reading on the abscissa of the latter by the corresponding reading on the abscissa of the former, then

$$\log x + \log \xi^2 = \log(r_0^2 \xi^2)$$

and in view of eq. (70) the above reduces to

$$r_0 = (\lambda_0/2\pi)x^{1/2} \quad (75)$$

To find the average particle radius, use is made of the above table and eqs. (72) and (75), thus

$$\bar{R} = (5x/3)^{1/2} (\bar{r}/r_0) (\lambda_0/2\pi) \quad (76)$$

where the ratio  $\bar{r}/r_0$  must be chosen for the particular value of  $n$  found in matching the curves. The curves drawn through the experimental points are the best matching theoretical curves.

#### PARTICLE SIZE DETERMINATIONS OF CARBON BLACKS

The matching process described above has yielded the following results for the two kinds of carbon black. See Figs. 12 and 13.

<u>Exposure Time</u> $\longrightarrow$	<u>3 Hrs. in H<sub>2</sub> Atmosphere</u>	<u>20 Hrs. in Air</u>
Carbon Black	$n = 0; x = 40 \times 10^4$	$n = 1; x = 10 \times 10^4$
54 Å radius	$\bar{R} = 45.5 \text{ Å}$	$\bar{R} = 69.4 \text{ Å}$



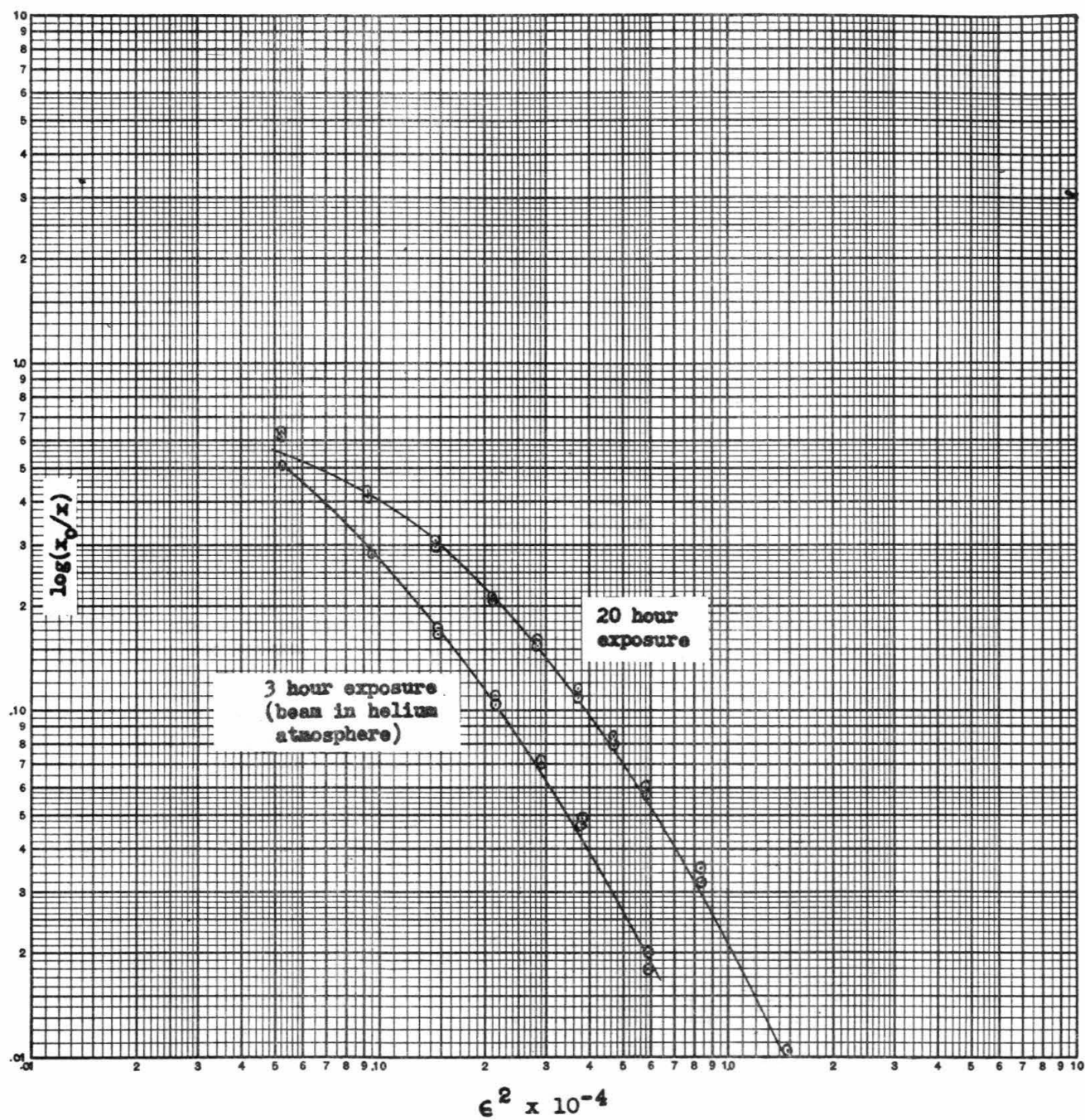


Fig. 12 - Carbon Black (Carbolac 1). Particle size - 50 Å radius.

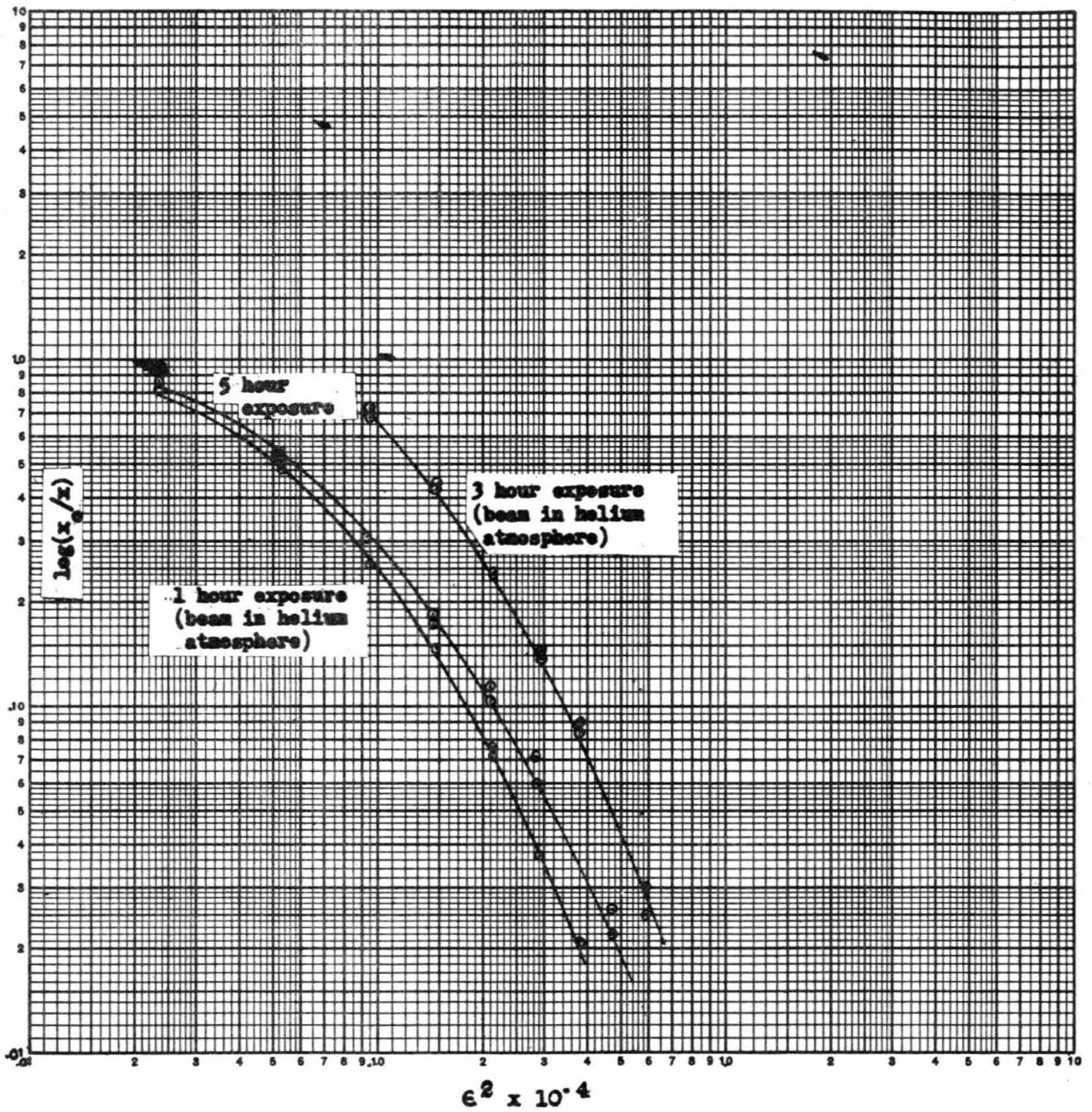


Fig. 13 - Carbon Black. Particle size - 230 Å radius.

Exposure Time	1 Hr. in He Atmosphere	5 Hrs. in Air	3 Hrs. in He Atmosphere
Carbon Black	$n=4; x=14.5 \times 10^4$	$n=3; x=14 \times 10^4$	$n=4; x=17.4 \times 10^4$
230 Å radius	$\bar{R} = 262 \text{ Å}$	$\bar{R} = 197 \text{ Å}$	$\bar{R} = 286 \text{ Å}$

Although the discrepancies in size determination for different exposures seem to be large at first sight, the actual experimental points, when shifted vertically to form one curve, fall in line quite nicely except for the end points which determine to a large extent the particular theoretical curve (and hence  $n$ ) to be used. The end points on the low intensity side of the experimental curves are hard to determine with accuracy because of the rather large grain size of ordinary x-ray film. The grains show up in the form of "grass" on the microphotometer curve. At the high intensity end, and hence at very small scattering angles, the discrepancy is probably due to either a slight amount of scattering by the tungsten wire (beam stop) or possibly incoherent scattering from the crystals, both of which may contribute to the scattered intensity at very low angles (i.e.  $\epsilon = 0.0015$  radians).

As one can see from the graphs, a fairly good exposure i.e. one that gives measurable results, can be obtained in 1 hour of exposure at 35PKV and 20 ma. with Helium atmosphere surrounding the beam. The direct beam can expose a film in about 15 seconds at 20 PKV and 20 ma. with air surrounding the beam. With the changes mentioned in Part III, this can be improved considerably.

REFERENCES

- (1) A. Guinier, Ann. de physique. (1939) 12, 161.
- (2) R.S. Bear and O.E.A. Bolduan, J.App. Phys.(1951) 22, 191.
- (3) R. von Nardroff, Phys. Rev. (1926) 28, 240.
- (4) C.G. Shull and L.C. Roess, J.App. Phys. (1947) 18, 295.
- (5) K.L. Yudowitch, J.App. Phys. (1949) 20, 174.  
(1949) 20, 1232.  
(1951) 22, 214.
- (6) O.E.A. Bolduan and R.S. Bear, J.App. Phys. (1949) 20, 983.
- (7) J.W.M. DuMond, R.S.I. (1950) 21, 188.
- (8) Compton and Allison, "X-ray in Theory and Experiment"  
Van Nostrand Co., Inc. New York (1935).
- (9) J. Johansson, Zeitschr. f. Phys. (1933) 82, 507.
- (10) H.H. Johann, Zeitschr. f. Phys. (1931) 69, 185.
- (11) J.E. White, J. App. Phys. (1950) 21, 855.
- (12) E. Ingelstam, Arkiv. f. Mat., Astron. och Fysik (1939)  
27B N:o 4.
- (13) J.W.M. DuMond, D.A. Lind, E.R. Cohen, R.S.I. (1947),  
18, 617.
- (14) P. Krishnamurti, Ind. J. Phys. (1930) 5, 472.
- (15) B. E. Warren, Phys. Rev. (1936) 49, 885.
- (16) J. Biscoe and B.E. Warren, J. App. Phys. (1942) 13, 364-371.

# PRELIMINARY RESULTS ON THE PARTICLE SIZE DETERMINATION OF LATEX SPHERES

Warren E. Danielson

This is a preliminary report on our work with the point-focusing monochromator in the determination of the mean diameter of latex spheres, and is included in this Special Technical Report to show, in an effective manner, the resolution of our instrument. This work is, in the future, to be expanded and incorporated with other applications of the instrument as part of a thesis in partial fulfillment of the requirements for the degree of Doctor of Philosophy at the California Institute of Technology.

The diffraction patterns of Dow latex (lot 3584, batch 580-G), consisting of nearly perfectly uniform spherical particles, were obtained by an experimental method similar to that used for the carbon-black measurements. The widespread interest in the size determination of these latex particles is emphasized by the fact that measurements by more than twenty investigators in three countries using at least six different methods have been reported. The values obtained for the diameter range from 2500 to 3200 Å with estimated accuracies ranging from  $\pm 25$  to  $\pm 240$  Å.\* Since these latex particles are used extensively in the calibration of electron microscopes, a more accurate knowledge of their size is of prime importance. An experiment similar in nature to the one we shall describe, but utilizing filtered chromium x-radiation and slit type focusing techniques has been reported by Yudowitch.\*\* The diffraction pattern obtained

---

\* C. H. Gerould, Journ. of Appl. Phys. 21, 183 (1950)

\*\* K. L. Yudowitch, Journ. of Appl. Phys. 22, 214 (1951)

by using uniform latex spheres as a scatterer consists in a series of concentric rings whose diameters can be easily measured with a microphotometer.

We believe that, for the purpose of size determination by x-ray diffraction, our instrument has a three-fold advantage over that used by Yudowitch. In the first place, fogging due to incoherent scattering by collimators is virtually eliminated. (In our case the "collimators" are the crystals themselves.) This is a result of the two successive Bragg reflections which the beam undergoes. The incoherent scattering of the continuous spectrum at the first crystal being completely suppressed at the second crystal. In the second place, our beam is highly monochromatic so that broadening of the diffraction rings due to inhomogeneity in the wavelength of the incident x-ray beam is negligible, and there is no uncertainty as to the wavelength to be used in interpreting the data. Finally, this highly monochromatic beam enables us to obtain several more measurable rings than were obtained by the above mentioned observer. The importance of these additional rings will be made clearer when we give an interpretation of the experimental results below. However, reference to equation 1 on page 86 at this point makes it clear that the values of the particle diameter which one computes from that equation are relatively less sensitive to the values,  $P$ , chosen for the packing fraction when  $u$  is large than when  $u$  is small.

#### EXPERIMENTAL RESULTS

Our sample consisted of latex from which the water of solution was removed by evaporation, the useful part of the sample being about  $3/4" \times 3/4" \times 0.1$  cm. We took several "pictures" of the diffraction



pattern, the exposure times varying from 1 hour to 80 hours. The two diffraction patterns reproduced on page 90 are typical of those we have obtained. (The dark oval spot near the center of each pattern is a series of punched round holes provided to allow the direct beam to pass through the film.) Successive rings, which are seen to be clearly resolved, are separated by a difference of scattering angle of about 0.002 radians. Page 91 shows a microphotometer trace of the 5 hour exposure. We tabulate the pertinent experimental data:

$d$  = Sample-to-film distance =  $642.8 \pm 0.5$  mm.

$\epsilon$  = Scattering angle.

$\epsilon d$  = Radius of diffraction ring produced at a scattering angle  $\epsilon$

Peak Number

Radius of ring, ( $\epsilon d$ ) mm.

(First peak measured is estimated with some uncertainty to be No. 5)	X-Ray Exposure Time					Average (Not weighted)
	1 hr.	5 hr.	23.7 hr.	41.6 hr.	80.7 hr.	
5	$1.80 \pm .04$	$1.83 \pm .01$				1.815
6	$2.15 \pm .03$	$2.20 \pm .01$	$2.18 \pm .02$			2.176
7		$2.56 \pm .02$	$2.53 \pm .02$			2.545
8		$2.93 \pm .01$	$2.90 \pm .02$	$2.92 \pm .03$	$2.91 \pm .02$	2.915
9		$3.30 \pm .02$	$3.28 \pm .02$	$3.29 \pm .02$	$3.29 \pm .02$	3.290
10		$3.70 \pm .03$	$3.63 \pm .03$	$3.66 \pm .01$	$3.66 \pm .02$	3.662
11		$4.04 \pm .03$	$4.03 \pm .02$	$4.03 \pm .02$	$4.03 \pm .02$	4.035
12			$4.40 \pm .02$	$4.40 \pm .01$	$4.40 \pm .03$	4.400
13				$4.76 \pm .02$	$4.77 \pm .02$	4.765
14					$5.14 \pm .02$	5.14
15					$5.51 \pm .02$	5.51

### INTERPRETATION OF RESULTS:

To interpret this pattern of diffraction rings and obtain from their measurement a value for the diameter of the latex particles, we have considered three possibilities:

(1) The spheres may tend to clump, in the process of drying, in such a way that each clump is made up of an hexagonal or cubic array of particles like a crystallite in a polycrystalline solid, the orientation of the clumps being random.

(2) The particles surrounding any arbitrarily chosen particle may fall into a spherically symmetric arrangement similar to that commonly assumed as representing atoms in a liquid.

(3) The particles could be treated as quite randomly distributed. Let us consider each of these possibilities separately.

One might suppose that electron microscope pictures of the latex such as those given by Gerould\* in the February 1950 issue of Journal of Applied Physics offer strong support for the first possibility. However, if this were the case in our sample, one would expect to find a series of rings (as in Debye-Sherrer powder patterns) corresponding to each fundamental spacing of "crystallite" planes containing a high particle surface density (i.e., one for each set of Miller indices of good reflecting power) and the fact that we have observed only one set of rings seems to render this "explanation" untenable.

To illustrate the analytical formulation of the second type of distribution suggested above, let us take the center of any one particle as the origin of a set of spherical polar coordinates and

---

\* C. H. Gerould, Journ. of Appl. Phys. 21, 185 (1950)



make some reasonable assumption for the number density of particles (i.e., particle centers) as a function of the radial distance from the origin. It is perhaps simplest (though certainly a rough approximation) to assume that the particle density is zero when the radius vector,  $r$ , is less than the particle diameter,  $D$ , that the density is highest for  $r = D$ , and that it has a somewhat lower (constant) value for  $r > D$ . This is precisely the assumption made by Yudowitch\* and it leads to a scattered intensity given by

$$I = MN^2 \bar{\Phi}^2(u) \left[ 1 + P \left\{ 5 \frac{\sin 2u}{2u} - 6 \bar{\Phi}(2u) \right\} \right] \quad (1)$$

where:

$I$  = x-ray intensity scattered at an angle  $\epsilon$  relative to that which would be scattered at the same angle by a single electron

$M$  = number of spheres irradiated in the sample

$N$  = number of electrons per sphere

$$\bar{\Phi}(u) = \frac{3}{u^3} (\sin u - u \cos u)$$

$$u = \frac{\pi \epsilon D}{\lambda}$$

$D$  = sphere diameter

$\lambda$  = x-ray wavelength

$P$  = degree of packing

The parameter  $P$ , called the packing fraction, is essentially a measure of the amount by which the particle density at  $r = D$  exceeds that for  $r > D$ . A plot of  $I$  vs.  $u$  reveals that a single set of rings (with relative ring diameters similar to those obtained experimentally) should be obtained if the theory is correct. A slightly different (and probably better) assumption for the spherical

---

\* Op cit.,

particle density, taking account of the lower density to be expected for values of  $r$  slightly greater than  $D$ , gives\*

$$I = MN^2 \overline{Q}^2 (u) \left[ 1 + P \left\{ 5 \frac{\sin 2u}{2u} - 6 \overline{Q} (2.36 u) \right\} \right] \quad (2)$$

which agrees with (1) except for the last term. The difference between the two equations is insignificant for the values of  $u$  which we consider. It should be noted that the predictions of equation (2) agree well with experiment in the case of scattering from liquid ether\* and that the particle size is small enough, in that case, to make the results at measurable angles sensitive to the term containing  $P$ .

The third assumption considered above, viz., that of random particle distribution, leads to an equation for the scattered intensity that is identical with (1) if  $P$  is taken equal to zero, so that again such an assumption is at least in qualitative agreement with the experimental results.

Assuming two different values for  $P$  in the first of the theoretical equations for the scattered intensity given above, and using the experimentally obtained values of  $\epsilon d$  as input data, we obtain the following results:

---

\* N. S. Gingrich and B.E. Warren, Phys. Rev. 46, 248 (1934)

$(\epsilon d)_{\text{ave}}$	$u_{\text{max}}$	Eqn. 1, $P = 0$		Eqn. 1, $P = 1/2$		
		$D(\text{\AA})$	$\text{Dev}(\text{\AA}) \equiv \delta_i$	$u_{\text{max}}$	$D(\text{\AA})$	$\text{Dev}(\text{\AA}) \equiv \delta_i$
1.815	15.515	2689	- 3	15.608	2705	+ 7
2.176	18.689	2702	+ 10	18.764	2712	+ 14
2.545	21.854	2701	+ 9	21.917	2709	+ 11
2.915	25.013	2699	+ 7	25.067	2705	+ 7
3.290	28.167	2693	+ 1	28.215	2698	0
3.662	31.320	2690	- 2	31.362	2694	- 4
4.035	34.470	2687	- 5	34.508	2690	- 8
4.400	37.619	2689	- 3	37.655	2692	- 6
4.765	40.767	2691	- 1	40.799	2693	- 5
5.14	43.914	2687	- 5	43.943	2689	- 9
5.51	47.060	2687	- 5	47.088	2688	- 10

$$D_{\text{ave}} = 2692 \text{ \AA}$$

$$\left( \frac{\sum_{i=1}^n \delta_i^2}{n(n-1)} \right)^{1/2} = 1.8 \text{ \AA}$$

$$D_{\text{ave}} = 2698 \text{ \AA}$$

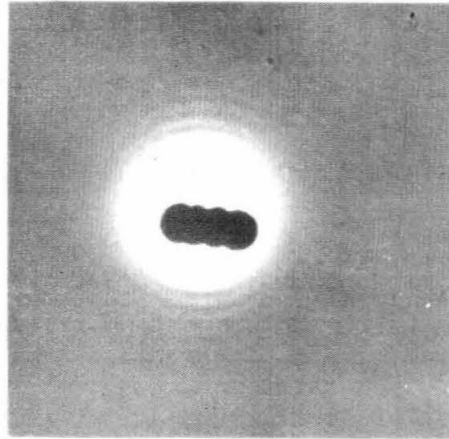
$$\left( \frac{\sum_{i=1}^n \delta_i^2}{n(n-1)} \right)^{1/2} = 2.6 \text{ \AA}$$

The values of the latex particle diameter obtained in the two cases differ by only  $6 \text{ \AA}$ , so that the effect of packing is seen to be rather small for the rings we have considered. Furthermore, the fact that there appears to be a decreasing trend in the diameters obtained from successively increasing ring sizes is probably significant and should be investigated in our further work. That  $2689 \text{ \AA}$  may be a better value for  $D$  than either of the averages given above is suggested by the fact that this is the average value

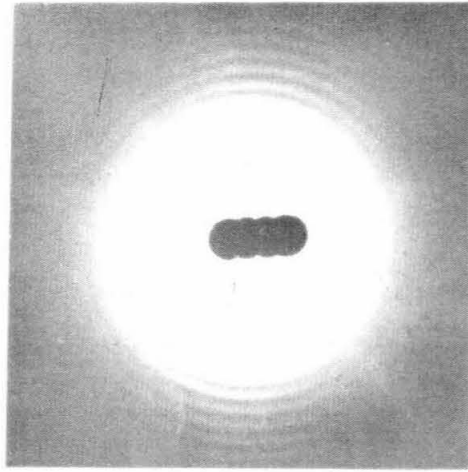
obtained from the last three lines in the table above, and the values are least sensitive to particle distribution for these higher values of  $u$ . The values of the diameter given above are also subject to slight change because of systematic errors, however, we believe such errors, other than the errors made in the assumptions underlying formulae (1) and (2), will effect the value of  $D$  by less than  $\pm 3 \text{ \AA}$ . The order of magnitude of the error introduced by inaccuracies in the theory can probably be inferred from the scatter in the values for  $D$  given in the table above.

In order to obtain more definite information as to the relative merits of the above assumptions, we propose: (1) to limit the size of the point focus by placing a pinhole with a diameter of about  $0.006''$  in front of the x-ray source, thereby making the "point-focus" small enough so that the presence or absence of a maximum in the pattern corresponding to a ring with a diameter of about  $2/3$  of a millimeter can be observed ( a packing fraction greater than about  $1/3$  predicts such a maximum while a smaller packing fraction does not); (2) to suspend the latex spheres in a liquid medium of high electron density (perhaps concentrated KI) and note whether the rings are still observable, and if so, whether the diameters have changed. The object of suspending the particles in a liquid is to ensure a sensibly random distribution. We expect these proposed experiments to be carried out in the near future.

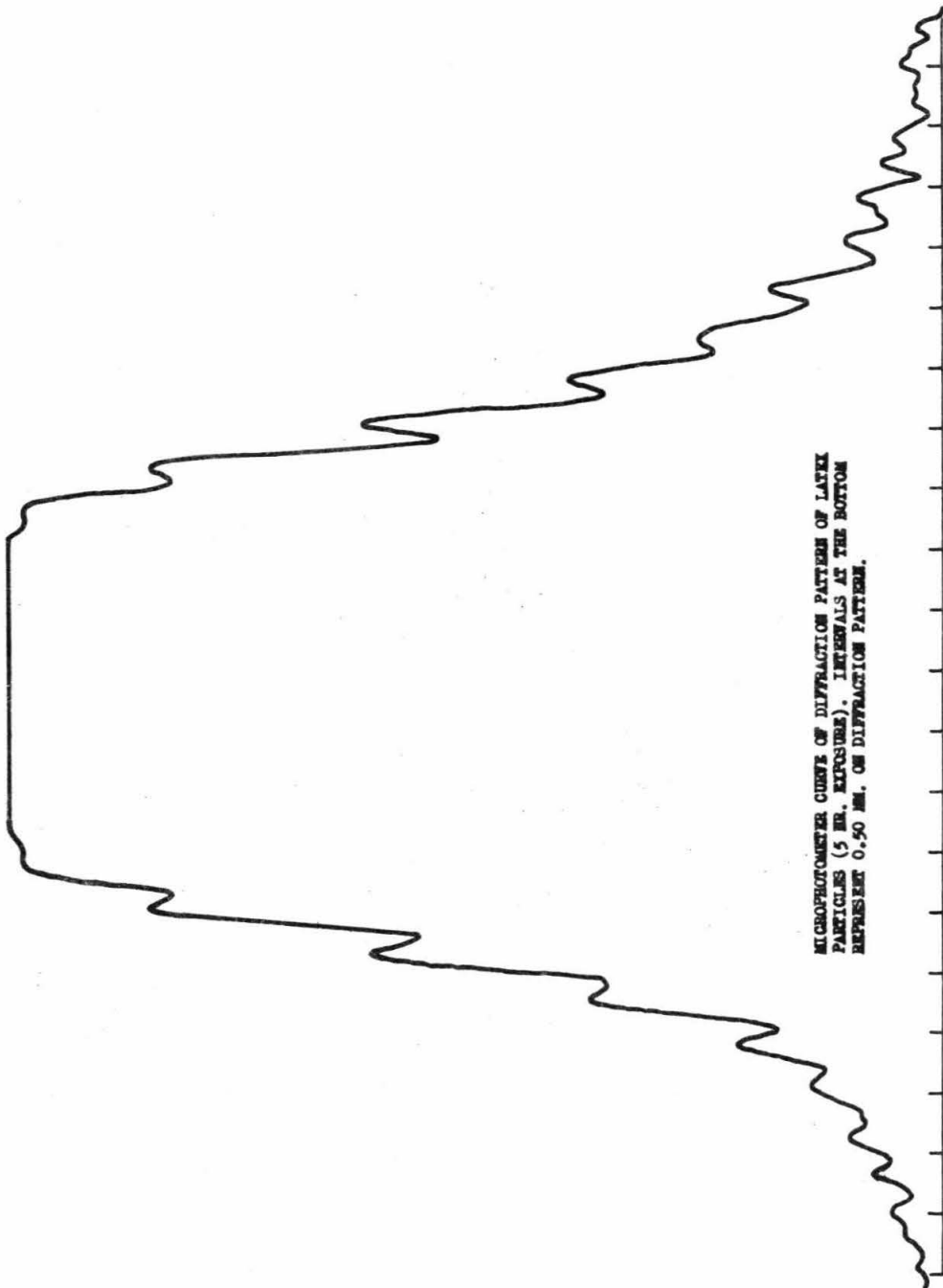
5 HR. EXPOSURE



80 HR. EXPOSURE



DIFFRACTION PATTERN OF LATEX SPHERES  
(APPROX. 2700 Å IN DIAMETER)



MICROPHOTOMETER CURVE OF DIFFRACTION PATTERN OF LATEX PARTICLES (5 HR. EXPOSURE). INTERVALS AT THE BOTTOM REPRESENT 0.50 MM. ON DIFFRACTION PATTERN.

TECHNICAL REPORTS DISTRIBUTION LIST

N6onr-244, T.O. IV  
NR 017-602

A. GOVERNMENT DISTRIBUTION

- |   |   |
|---|---|
| (2) Research & Development Board<br>Committee on Electronics<br>Pentagon Bldg., 1712 G.St., N.W.<br>Washington 25, D.C.             | (2) Office of Assist. Naval Attache<br>for Research<br>Navy - 100<br>Fleet Post Office<br>New York, N.Y.  |
| (2) Chief of Naval Research<br>Office of Naval Research<br>Washington 25, D.C.<br>Attn: Physics Branch                              | (1) Chief of Bureau of Ordnance<br>Navy Department<br>Washington 25, D.C.<br>Attn: Rea  |
| (9) Director, Naval Research Lab.<br>Washington 20, D.C.<br>Attn: Tech. Info. Officer   | (1) Chief, Bureau of Ships<br>Navy Department<br>Washington 25, D.C.<br>Attn: Code 330  |
| (1) Commanding Officer<br>U.S. Navy O.N.R.<br>495 Summer Street<br>Boston 10, Massachusetts   | (1) Chief of Bureau of Aeronautics<br>Navy Department<br>Washington 25, D.C.<br>Attn: AE-4  |
| (1) Commanding Officer<br>U.S. Navy O.N.R.<br>Branch Office<br>50 Church Street<br>New York 7, N.Y.                                 | (1) Commanding Officer<br>Engineering Research & Develop. Lab<br>Fort Belvoir, Virginia   |
| (1) Commanding Officer<br>U.S. Navy O.N.R.<br>Branch Office<br>American Fore Bldg.<br>844 North Rush Street<br>Chicago 11, Illinois | (1) Office of the Chief of Ordnance<br>Research & Develop. Service<br>Department of the Army<br>The Pentagon<br>Washington 25, D.C.<br>Attn: ORDTB-Research Coordination<br>Br. |
| (1) Commanding Officer<br>U.S. Navy O.N.R.<br>Branch Office<br>801 Donahue Street<br>San Francisco 24, Calif.                       | (1) Commanding General<br>Air Materiel Command<br>Wright-Patterson Air Force Base<br>Dayton, Ohio   |
| (1) Commanding Officer<br>U.S. Navy O.N.R.<br>Branch Office<br>1030 E. Green Street<br>Pasadena 1, California                       | (1) Head, Division P, Los Alamos Nat'l<br>Lab., Los Alamos, New Mexico  |
| (1) Director, Naval Research Lab.<br>Washington 20, D.C.<br>Attn: Code 2021   | (1) Nat'l Advisory Comm. for Aeronautics<br>Lewis Flight Propulsion Lab.<br>Cleveland Airport<br>Cleveland 11, Ohio   |
|   | (1) National Research Council<br>Division of Physical Sciences<br>National Academy of Sciences<br>Washington 25, D.C.   |

- |  |  |
|--|--|
| (1) Director, Information Div.<br>Argonne Nat'l Laboratory<br>Chicago 80, Illinois                             | (1) Chief, Isotopes Division<br>Oak Ridge Operations<br>Post Office Box "E"<br>Oak Ridge, Tennessee<br>Attn: Dr. Paul C. Aebersold |
| (1) Director, Information Division<br>Brookhaven Nat'l Laboratory<br>Upton, Long Island, New York              | (1) Atomic Energy Commission<br>Division of Research<br>Washington 25, D.C.<br>Attn: Dr. J. Platt                                  |
| (1) National Bureau of Standards<br>Central Radio Propagation Lab.<br>Washington 25, D.C.<br>Attn: R.C. Peavey |  |

B. NON-GOVERNMENT DISTRIBUTION

- |   |  |
|---|--|
| (1) California Institute of Technology<br>Pasadena, California<br>Attn: Linus Pauling   | (1) State College of Washington<br>Department of Physics<br>Pullman, Washington<br>Attn: Dr. S. Town Stephenson    |
| (1) Laboratory for Insulation Research<br>Mass. Institute of Technology<br>Cambridge 39, Massachusetts<br>Attn: Dr. A. Von Hippel | (1) University of Illinois<br>Department of Physics<br>Urbana, Illinois<br>Attn: Dr. F. Seitz                      |
| (1) Brooklyn Polytechnic Institute<br>Department of Physics<br>Brooklyn, N.Y.<br>Attn: Dr. I. Fankuchen                           | (1) University of Wisconsin<br>Department of Physics<br>Madison, Wisconsin<br>Attn: Dr. W.V. Beeman                |
| (1) Carnegie Institute of Technology<br>Department of Physics<br>Pittsburgh, Pennsylvania   | (1) Ohio State University Research<br>Foundation<br>Physics Department<br>Columbus 10, Ohio<br>Attn: Dr. C.H. Shaw |
| (1) Attn: Dr. E. Teller<br>(1) Dr. C. Zener<br>University of Chicago<br>Department of Physics<br>Chicago, Illinois                | (1) Professor S.K. Allison<br>Institute for Nuclear Studies<br>University of Chicago<br>Chicago, Illinois          |
| (1) Dr. Lyman G. Parratt<br>Dr. Lloyd F. Smith<br>Cornell University<br>Department of Physics<br>Ithaca, N.Y.                     | (1) University of California<br>Department of Physics<br>Berkeley, California<br>Attn: Dr. Luis W. Alvarez         |
| (1) Research Laboratory of Electronics<br>Mass. Institute of Technology<br>Cambridge 39, Massachusetts<br>Attn: Dr. J.A. Stratton | (1) Dr. J. A. Bearden<br>Johns Hopkins University<br>Department of Physics<br>Baltimore, Maryland                  |



- (1) Prof. H. A. Bethe  
Physics Department  
Cornell University  
Ithaca, New York
- (1) Dr. Felix Bloch  
Department of Physics  
Stanford University  
Palo Alto, California
- (1) Prof. A. H. Compton  
Washington University  
St. Louis, Mo.
- (1) Dr. James M. Cork  
University of Michigan  
Ann Arbor, Michigan
- (1) Dr. Ludo K. Frevel  
2-294E  
Dow Chemical Company  
Midland, Michigan
- (1) Dr. J. E. Henderson  
University Of Washington  
Seattle, Washington
- (1) Prof. A. L. Hughes  
Physics Department  
Washington, University  
St. Louis, Mo.
- (1) ~~Dr. P. Kusch~~  
Princeton University  
Princeton, New Jersey
- (1) Dr. P. Kusch  
Physics Department  
Columbia University  
New York, New York
- (1) Prof. Raymond Birge  
University of California  
Department of Physics  
Berkeley, California
- (1) Dr. Martin Deutsch  
Mass. Institute of Technology  
Department of Physics  
Cambridge 39, Mass.
- (1) Dr. L. Jackson Laslett  
Institute for Atomic Research  
Iowa State College  
Ames, Iowa
- (1) Dr. E. O. Lawrence  
Department of Physics  
University of California  
Berkeley, California
- (1) Dr. C. E. Mandeville  
Bartol Research Foundation  
Swarthmore, Pennsylvania
- (1) Dr. Allan C. G. Mitchell  
Physics Department  
University of Indiana  
Bloomington, Indiana
- (1) Dr. Edwin McMillan  
Physics Department  
University of California  
Berkeley, California
- (1) Dr. J. Robert Oppenheimer  
Institute for Advanced Study  
Princeton, New Jersey
- (1) Prof. I.I. Rabi  
Physics Department  
Columbia University  
New York City, N.Y.
- (1) Dr. L. I. Schiff  
Physics Department  
Stanford University  
Palo Alto, California
- (1) Prof. John A. Wheeler  
Palmer Physical Laboratory  
Princeton University  
Princeton, N.J.

OPTIONAL DISTRIBUTION LIST

Spec. Tech. Rep. No. 15

- |  |   |
|--|---|
| (1) Director, Nat'l Bureau of Standards<br>Washington, D.C.  | (1) Director, Information Division<br>U.S. Atomic Energy Commission<br>Washington 25, D.C.                |
| (1) Commanding Officer, U.S. Navy O.N.R.<br>Branch Office<br>New York Naval Shipyard<br>Brooklyn 1, New York                   | (1) Director, Naval Research Laboratory<br>Anacostia Station<br>Washington 25, D.C.                       |
| (1) Chief of Naval Operations<br>Navy Department<br>Washington, D.C.   | (1) Nat'l Bureau of Standards, Library<br>Room 203<br>N.W. Bldg.<br>Washington 25, D.C.                   |
| (1) Director<br>Naval Ordnance Laboratory<br>Naval Gun Factory<br>Washington 25, D.C.<br>Attn: L.H. Rumbaugh                   | (1) Director,<br>Dept. of Terrestrial Magnetism<br>Carnegie Institution of Washington<br>Washington, D.C. |
| (1) Chief,<br>Eng. & Tech. Div., SIGTM-S<br>Office of the Chief Signal Officer<br>The Pentagon<br>Washington, D.C.             | (1) Director<br>Bartol Research Foundation<br>Franklin Institute<br>Swarthmore, Pa.                       |
| (1) National Institute of Health<br>Public Health Nat'l Sec. Agency<br>Bethesda 14, Maryland<br>Attn: Dr. Albert Szent Gyorgyi | (1) Radiation Laboratory Director<br>University of California<br>Berkeley, California                     |
| (1) Director, Research Laboratories<br>Westinghouse Electric Corporation<br>East Pittsburgh, Pa.                               | (1) Director, Inst. of Nuclear Studies<br>University of Chicago<br>Chicago, Illinois                      |
| (1) Director, Research Laboratory<br>General Electric Corporation<br>Schenectady, New York                                     | (1) Backus, R.C.<br>Research Assoc. in Biophysics<br>University of Michigan<br>Ann Arbor, Michigan        |
| (1) Officer in Charge, Radiation Lab.<br>San Francisco Naval Shipyard<br>San Francisco, California                             | (1) Bauer, Dr. H.E.G.<br>11 Rue Pierre Curie<br>Laboratoire de Chimie Physique<br>Paris, France           |
| (1) Director, Research Division<br>U.S. Atomic Energy Commission<br>Washington 25, D.C.  | (1) Bear, Dr. Richard S.<br>Biology Department<br>Mass. Inst. of Technology<br>Cambridge 39, Mass.        |
| (1) Head, Physics Division<br>Clinton National Laboratory<br>Oak Ridge, Tennessee  | (1) Barrett, Charles S.<br>Inst. for the Study of Metals<br>University of Chicago<br>Chicago, Illinois    |

OPTIONAL DISTRIBUTION

-5-

- (2) Bragg, Sir Lawrence  
Cavendish Laboratory  
Cambridge, England
- (1) Burgers, Prof. H.C.  
Laboratory for Physical Chemistry  
136 Julianalaan  
Delft, Holland
- (1) Bolduan, Orville E.A.  
Camp Detrick  
Frederick, Maryland
- (1) Cauchois, Mlle. Yvette  
Laboratoire de Chimie Physique  
11 Rue Pierre Curie  
Paris 5, France
- (1) California Research Corp., P.O. Box 446  
La Habra, California  
Attn: Director
- (1) Coster, Dirk  
Universitet Gröningen  
Natuurkundig Laboratorium  
Gröningen, Netherlands
- (1) Christy, Prof. R. F.  
California Inst. of Tech.  
Pasadena, California
- (1) Daniel, Dr. Vera  
Electrical Research Assoc.  
5 Wadsworth Road  
Greenford-Middlesex, England
- (1) Dandliker, W.B.  
Harvard Medical School  
Cambridge, Mass. (Forward)
- (1) Darwin, Dr. C.G.  
National Physical Laboratories  
Teddington, England
- (1) Déjardin, Prof. G. L.  
Faculté des Sciences  
Inst. de Physique Générale  
18 Quai Claude-Bernard  
Lyon-Rhone, France
- (1) De Wolff, Dr. P.M.  
Laboratory for Technical Physics  
Delft, Holland
- (1) Faessler, Dr. A.  
Katharinenstr. 25  
Freiburg i, Br. Germany
- (1) Fowler, Prof. William  
Calif. Institute of Tech.  
Pasadena, California
- (2) Farrant, J.L., A.J. Hodge  
Australian Council for Scientific  
& Industrial Research  
Australia
- (1) Godfrey L. Cabot, Inc.  
49 Beach St.,  
Boston 11, Massachusetts  
Attn: M. Martin
- Fullam E. F.  
(1) Knolls Research Laboratory  
General Electric Co.  
Schenectady, New York
- (1) Garner, Clifford  
Chemistry Department  
U.C.L.A.  
Los Angeles, 24, California
- (1) Glocker, Prof. R.  
Röntgeninstitut der Tech.Hochschule  
Stuttgart N.  
Württ, Deutschland  
U.S.A. Zone
- (1) Gerould, C.H.  
Dow Chemical Co.  
Magnesium Laboratories  
Midland, Michigan
- (1) Gosting, Mr. Louis  
Rockefeller Inst. for Med. Research  
66th St. and York Avenue  
New York 21, New York
- (1) Grimmett, Dr. L.G.  
University of Texas  
M.D. Anderson Hospital  
Houston, Texas
- (1) Gueben, Dr. G.  
Université de Liège  
Institut de Physique  
1 A, quai Roosevelt  
Liege, Belgium

OPTIONAL DISTRIBUTION

-6-

- (2) Guinier, Dr. André J.  
44 Boulevard Port Royal  
Paris 5, France
- (1) Hayashi, Dr. Takeshi  
Inst. of Physics  
Faculty of Science  
Tohoku University  
Sendai, Japan
- (1) Ewald, P. P.  
Brooklyn Polytechnic Institute  
Brooklyn, New York
- (1) Hellner, Dr. E.  
Mineralogisches Inst. der Universität  
Deutschhausstr. 10  
Marburg, Germany
- (1) Houston, Dr. W.V.  
Rice Institute of Technology  
Houston, Texas
- (1) Hughes, Prof. E.W.  
Chemistry Department  
California Inst. of Tech.  
Pasadena, California
- (1) Ingelstam, Dr. Eric  
Institutionen f. Fysik  
Kungl. Techniska Hogskolan  
Stockholm 26, Sweden
- (2) Kern, Dr. S. F.  
Kern, Dr. R.A.  
Eli Lilly Co.  
Indianapolis, Indiana
- (1) Kahler, Dr. Herbert  
Nat'l Inst. of Health  
Bethesda, Maryland
- (1) Kirkpatrick, Dr. Paul  
Physics Department  
Stanford University  
Palo Alto, California
- (1) Linde Air Products Co., Gen. Res. Lab.  
East Park Drive & Woodward Avenue  
Tonawanda, New York  
Attn: A.C. Jenkins
- (1) Lauritsen, Dr. C. C.  
California Inst. of Tech.  
Pasadena, California
- (1) Lind, Dr. David A.  
University of Wisconsin  
Madison, Wisconsin
- (1) Lonsdale, Dr. K.  
University College  
Gower Street  
London W.C. 1, England
- (1) Massachusetts Institute of Technology  
Research Lab. of Electronics  
Cambridge 39, Massachusetts  
Attn: John Hewitt - Document Office
- (1) Magat, Dr.  
Laboratoire de Chimie Physique  
11 Rue Pierre Curie  
Paris, France
- (1) Marton, Dr. Louis  
National Bureau of Standards  
Washington 25, D.C.
- (1) Millikan, Dr. Robert A.  
Physics Department  
Calif. Inst. of Tech.  
Pasadena, California
- (1) Nijgh, Dr. G. J.  
Zeeman Laboratorium  
Harwell, England
- (1) North, Mr. Philip  
Dow Chemical Co.  
Midland, ~~Washington~~ Michigan
- (1) North American Aviation Corp.  
A.E. Research Department Library  
Downey, California
- (1) Panofsky, Prof. W.K.H.  
Department of Physics  
Stanford University  
Stanford, California
- (1) Prins, Dr. J.A.  
Laboratory for Technical Physics  
Delft, Holland

OPTIONAL DISTRIBUTION

-7-

- (1) Rathenau, Mr. G.  
Philips Research Laboratory  
Eindhoven, Holland
- (1) Rossini, Dr. Fred D.  
Chm., Nat'l Research Council  
Carnegie Institute of Technology  
Schenley Park  
Pittsburgh 13, Pa.
- (1) Richards, Dr. O.W.  
American Optical Co.  
Buffalo 15, N.Y.
- (1) Roess, L.C.  
Beacon Research Lab. of Texas Co.  
P.O. Box 509  
Beacon, New York
- (1) Rogossa, Dr. George  
Florida State University  
Tallahassee, Florida
- (1) Scherrer, Prof. P.  
Technische Hochschule  
Zurich, Switzerland
- (1) Shell Development Co.  
Emeryville, California  
Attn: Director
- (1) Scott, G.D.  
University of Toronto  
Toronto, Ontario, Canada
- (1) Seitz, Prof. F. D.  
Carnegie Inst. of Technology  
Pittsburgh, Pa. (Forward)
- (1) Siegbahn, Dr. Manne  
Nobelinstitut f. Fysik  
Stockholm 50, Sweden
- (1) Sharp, D. G.  
Department of Biophysics  
Duke University  
Durham, North Carolina
- (1) Stille, Dr. H. U.  
Physikalische Technische  
Reichsanstalt  
Braunschweig, Cyriaksring 43,  
Germany
- (1) Shull, C.G.  
Clinton Laboratories  
Oak Ridge, Tennessee
- (1) Spencer, R.S.  
Dow Chemical Co.  
Midland, Michigan
- (1) Trillat, Prof. J.J.  
Lab. de Bellevue C.N.R.S.  
8 Rue Jeanne d'arc  
St. Germain-en-Laye  
(Seine et Oise) France
- (1) Verster, Dr. N.F.  
Zeeman Laboratorium  
Harwell, England
- (1) Von Laue, Dr. Max  
Gottingen  
Bunsenstrasse 16  
British Zone, Germany
- (1) Warren, Prof. B.E.  
Department of Physics  
Mass. Inst. of Tech.  
Cambridge 39, Mass.
- (1) Williams, R. C.  
University of Michigan  
Ann Arbor, Michigan
- (1) Yudowitch, Dr. K.L.  
Department of Physics  
Florida State University  
Tallahassee, Florida
- (1) Zachariason, Dr. W.F.H.  
University of Chicago  
Chicago, Illinois
- (1) Schwarz, Dr. G.  
Department of Physics  
Florida State University  
Tallahassee, Florida
- (1) Watson, Dr. Bernard B.  
Federal Security Agency  
Department of Education  
Washington, D.C.
- (1) B.F. Goodrich Co.  
Research Laboratory  
Brecksville, Ohio  
Attn: E.H. Rowe

OPTIONAL DISTRIBUTION

-8-

- |   |   |
|---|---|
| (1) Bacher, Dr. Robert<br>Chm., Physics Department<br>Calif. Inst. of Tech.<br>Pasadena, California | (1) Princeton University<br>Princeton, New Jersey   |
| (1) Corey, Prof. R. B.  | (1) Purdue University<br>Lafayette, Indiana   |
| (1) Sturdivant, Prof. J. H.   | (1) Rice Institute<br>Houston, Texas  |
| (1) Yost, Prof. D.<br>Department of Chemistry<br>Calif. Inst. of Tech.<br>Pasadena, California      | (1) Sloane Physics Laboratory<br>Yale University<br>New Haven, Connecticut                    |
| (1) Beadle, Prof. G.W.  | (1) Stanford University<br>Palo Alto, California  |
| (1) Borsook, Prof. H.   | (1) New York University<br>Washington Square College<br>Washington Square<br>New York 3, N.Y. |
| (1) Delbruck, Prof. M.  | (1) Washington University<br>St. Louis, Mo.   |
| (1) Weigle, Prof. J.  | (1) University of California<br>Berkeley, California  |
| (1) Wiersma, Prof. C.A.G.<br>Department of Biology<br>Calif. Inst. of Tech.<br>Pasadena, California | (1) University of California<br>Los Angeles 24, California                                    |
| <u>Chairman, Department of Physics of:</u>  |   |
| (1) University of Notre Dame<br>Notre Dame,<br>Indiana  | (1) University of Chicago<br>Chicago 37, Illinois   |
| (1) Catholic University of America<br>Washington 17, D.C.   | (1) University of Colorado<br>Boulder, Colorado   |
| (1) Columbia University<br>New York 27, New York  | (1) University of Illinois<br>Urbana, Illinois  |
| (1) Duke University<br>Durham, North Carolina   | (1) State University of Iowa<br>Iowa City, Iowa   |
| (1) Harvard University<br>Cambridge, Massachusetts  | (1) University of Kansas<br>Lawrence, Kansas  |
| (1) Indiana University<br>Bloomington, Indiana  | (1) University of Minnesota<br>Minneapolis, Minnesota   |
| (1) Iowa State College<br>Ames, Iowa  | (1) University of Michigan<br>Ann Arbor, Michigan   |
| (1) Johns Hopkins University<br>Baltimore, Maryland   | (1) University of Pittsburgh<br>Pittsburgh, Pa.   |
| (1) Northwestern University<br>Evanston, Illinois   |   |
| (1) Ohio State University<br>Columbus 10, Ohio  |   |

OPTIONAL DISTRIBUTION

-9-

- (1) University of Pennsylvania  
Philadelphia, Pa.
- (1) University of Rochester  
Rochester, New York
- (1) University of Southern California  
Los Angeles, California
- (1) University of Washington  
Seattle;5, Washington
- (1) University of Wisconsin  
Madison 6, Wisconsin
- (1) Rutgers University  
New Brunswick, New Jersey
- (1) Director  
Applied Physics Laboratory  
Johns Hopkins University  
Silver Spring, Maryland
- (1) New York University  
University Heights  
New York 53, New York